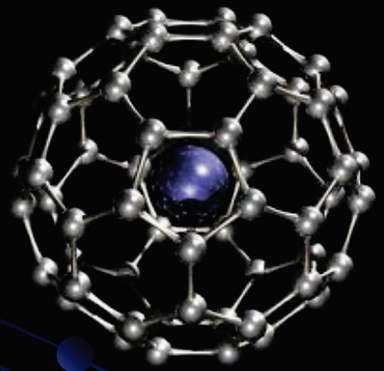
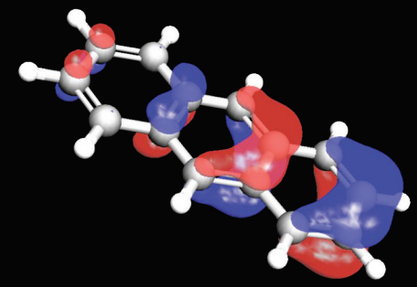
TDDFT winter school - Benasque january 2012

Physics with Short Laser Pulses





Franck Lépine

LASIM - Lyon (France)

Physics with short pulses

-I- Basics

-II- An example of attosecond experiments:
Electron localization

-III- Other examples
Transient attosecond processes

-IV- Rabbit & Streaking
Shearing

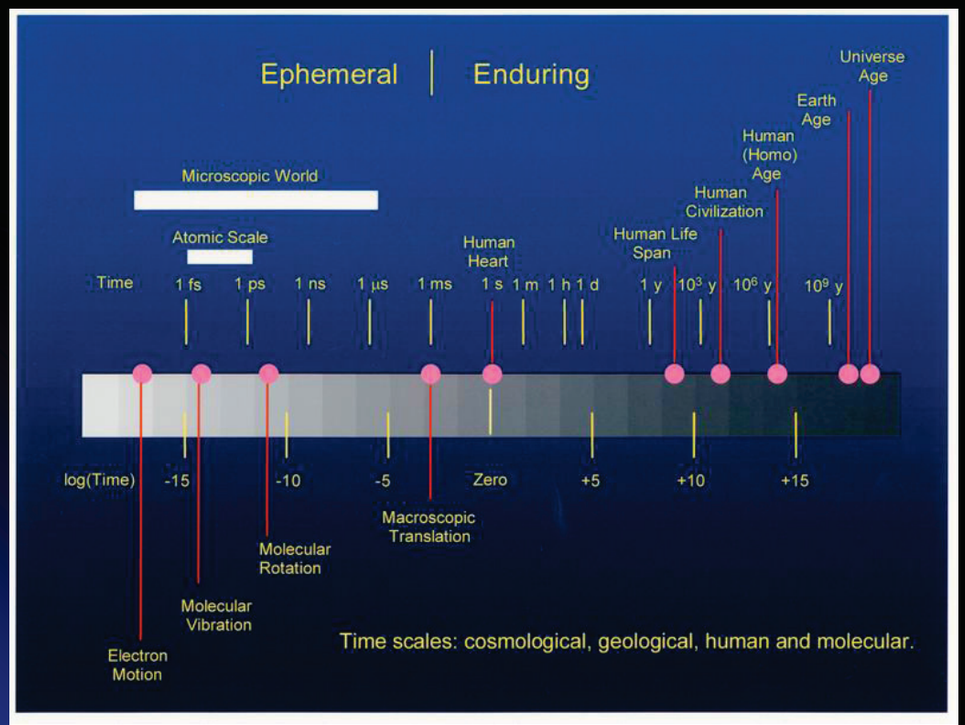
-V- Delay in photoemission

-VI-strong field holography

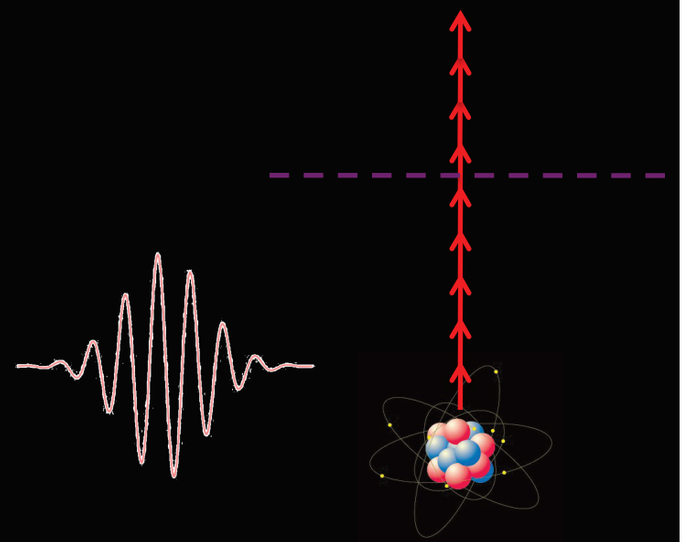
-I- Basics

Short Pulses… What for?

- Ultrafast dynamics



- Strong field
/ non-linear photophysics



Short Pulses… What for?

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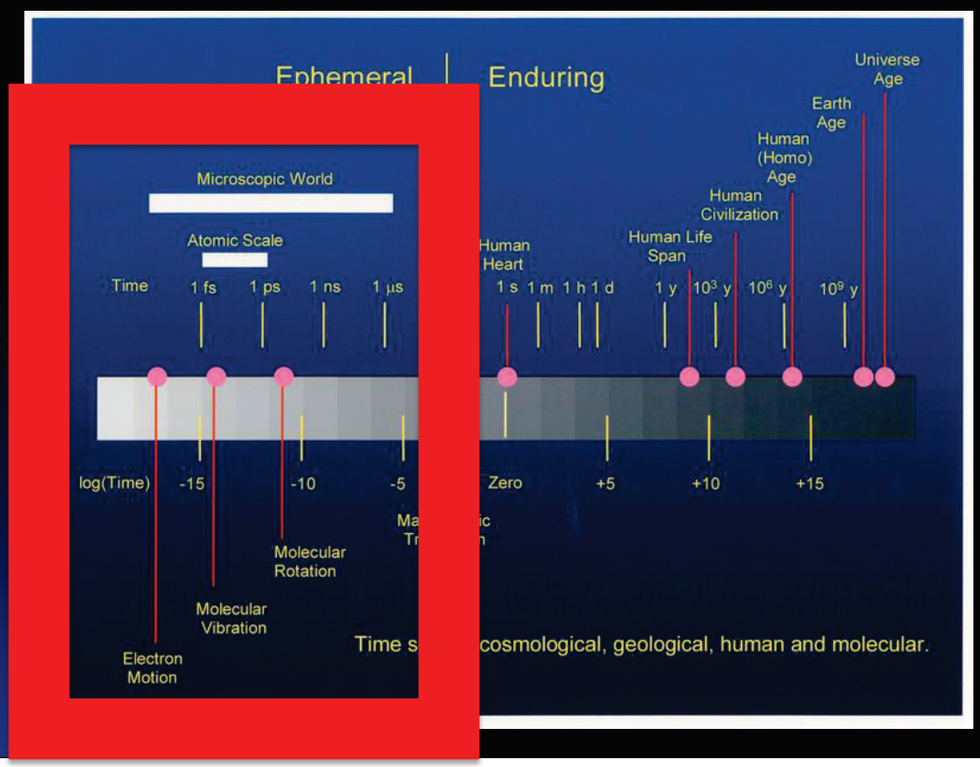

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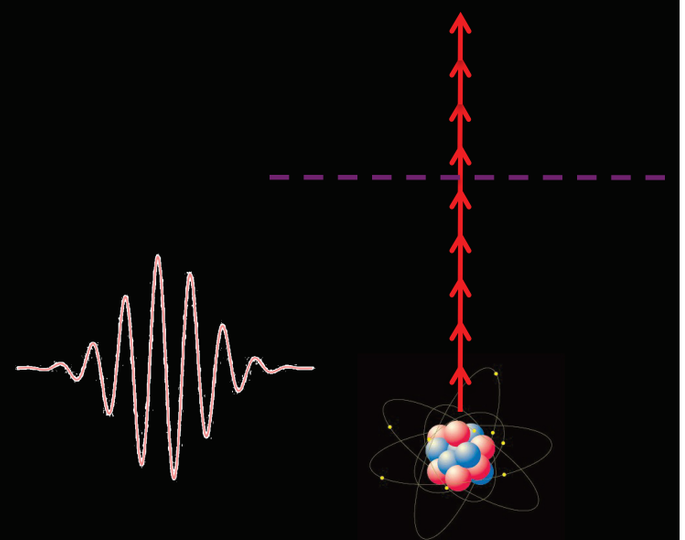

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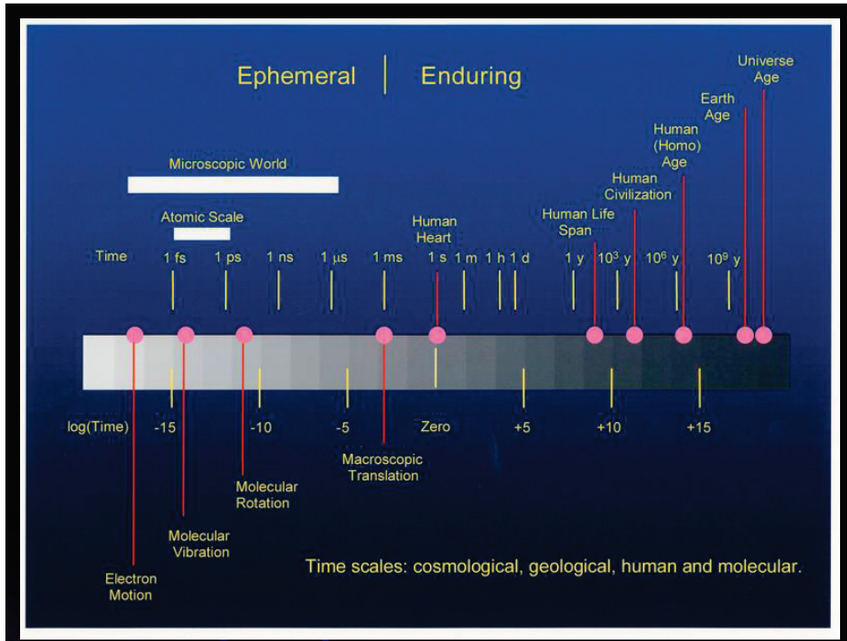
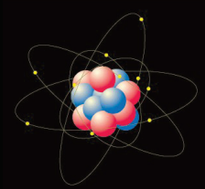


How Short ?

=> femtosecond … attosecond timescale



τ_{atomic} = 24 attoseconds

« femtosecond » Timescale of the nuclear motion (fs-chemistry)

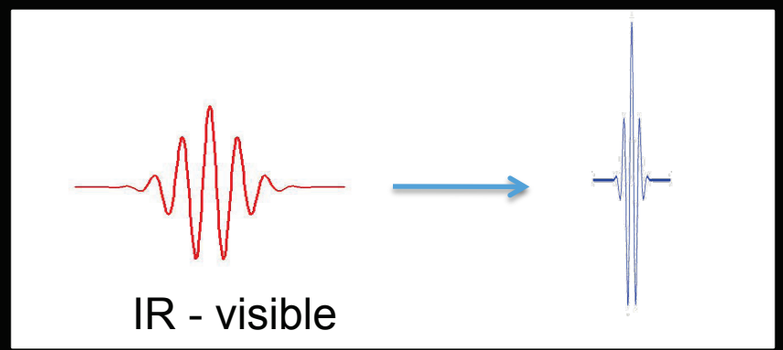
« attosecond » Timescale of the electron motion on the atomic scale

Modern Light Sources

recent progresses ….

- Shorter and shorter pulses down to the attosecond timescale

High harmonic generation sources



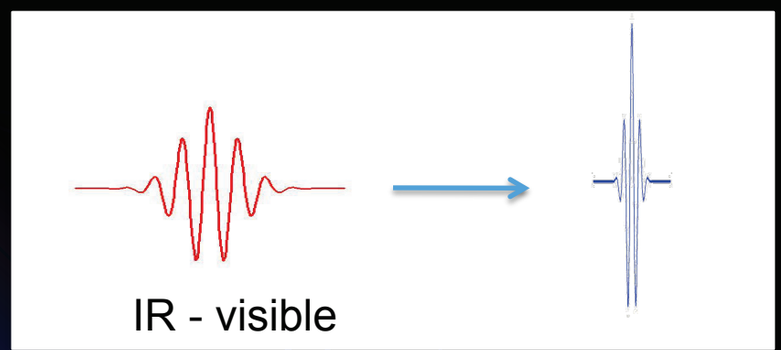
- More Intense light pulses for extreme wavelength (*Far IR, VUV, XUV etc…)*

Free electron lasers (LCLS, FLASH, FELICE …)

Modern Light Sources

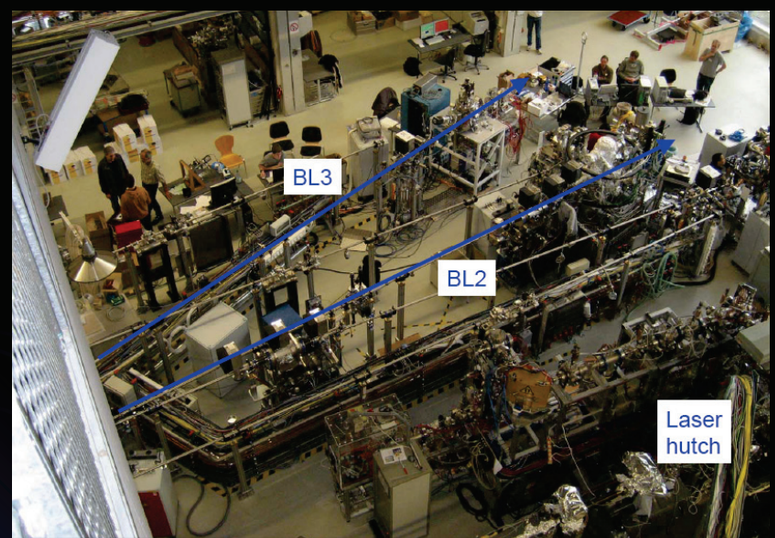
Changing light properties …

High harmonic generation source



Modern Light Sources

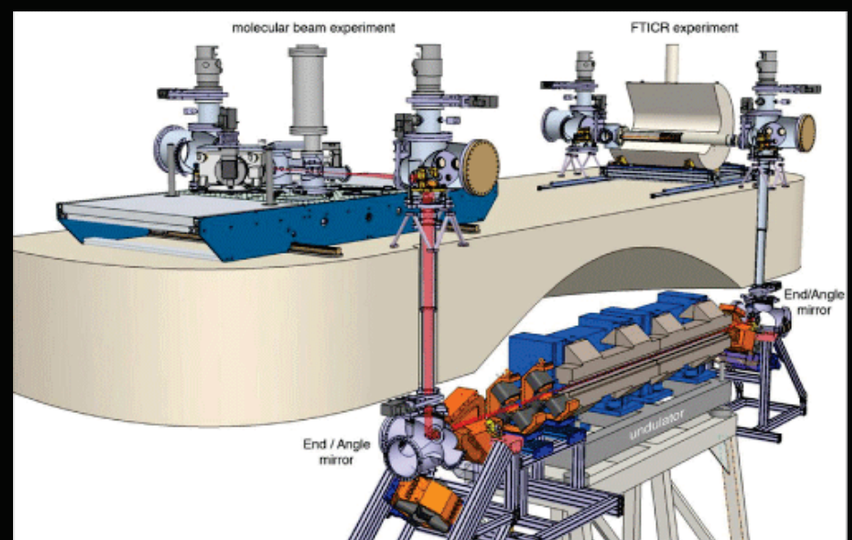
Changing light properties …



FLASH (Hamburg)

XUV

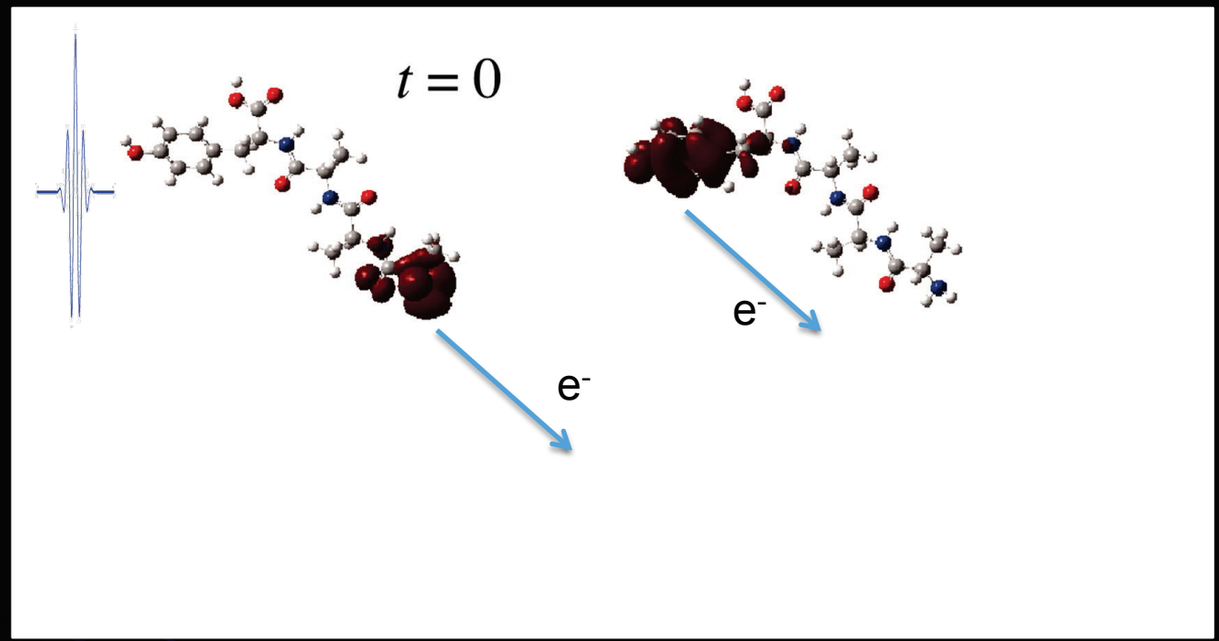
FREE ELECTRON LASERS



FELICE user's facility (The Netherlands)

Far IR

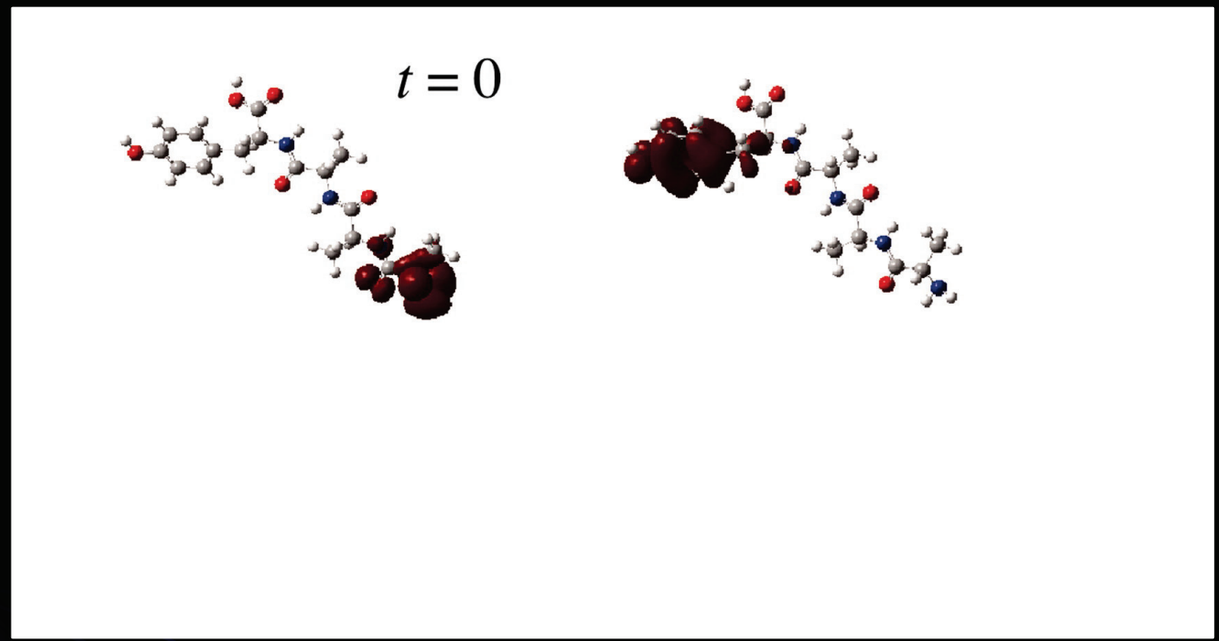
Hole propagation in peptides following photoionization



Snapshots of the densities of the hole as a function of time for TyrAla3 for ionization of the HOMO (Left) and HOMO-1 (Right).

Remacle F , Levine R D, PNAS 2006;103:6793-6798

Hole propagation in peptides following photoionization



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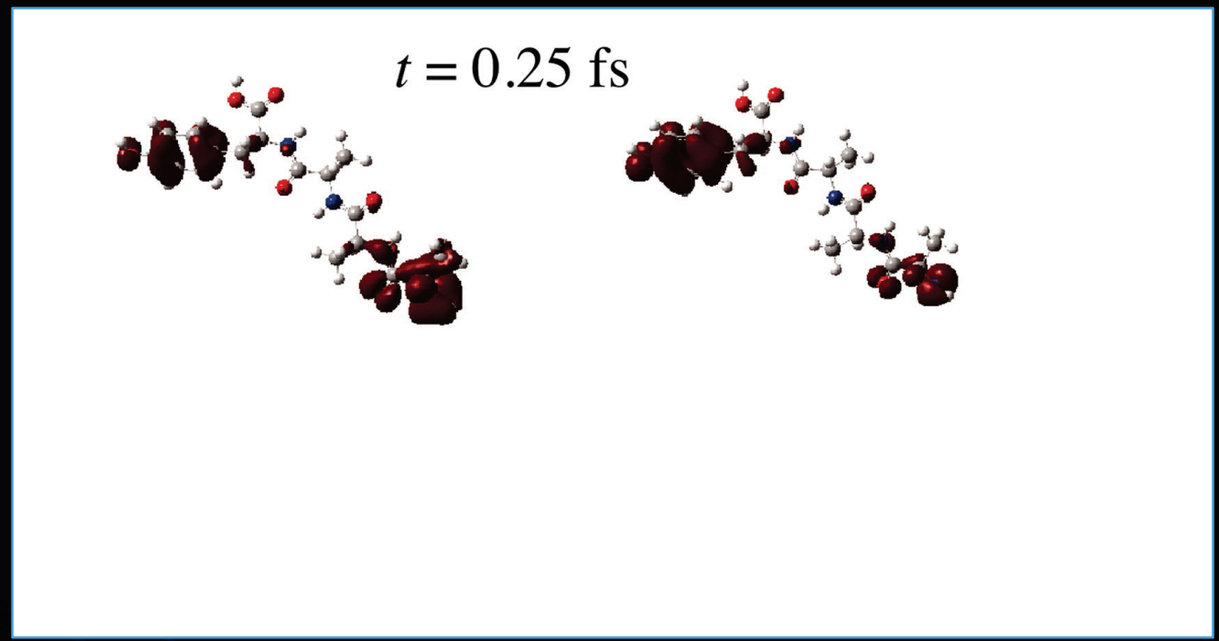

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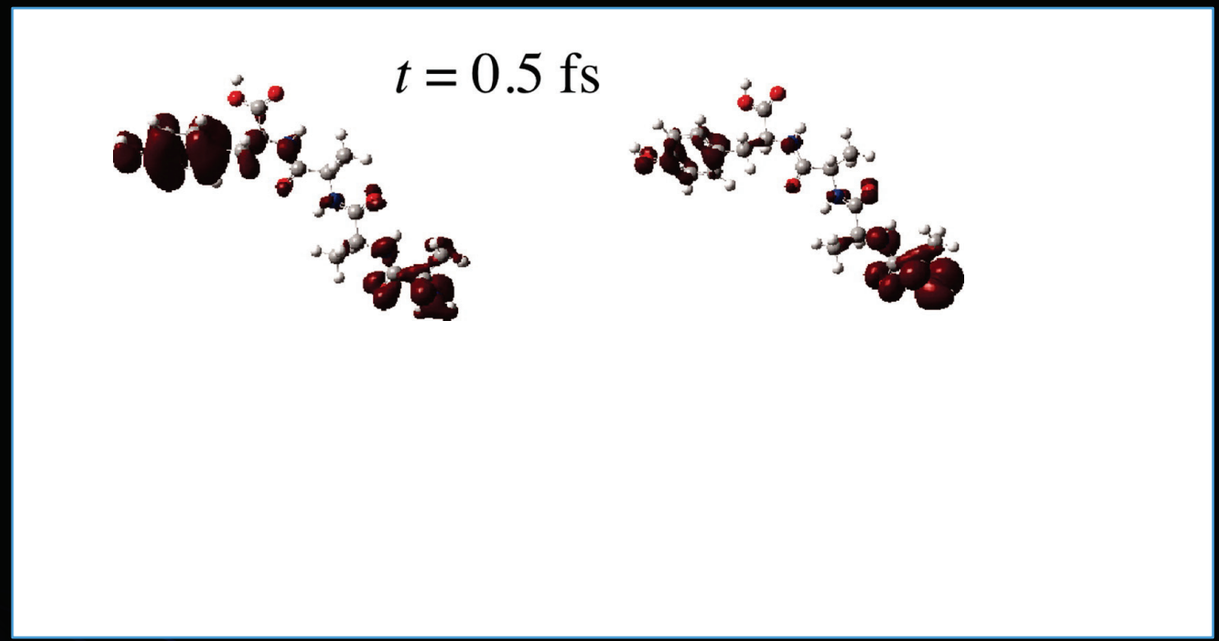

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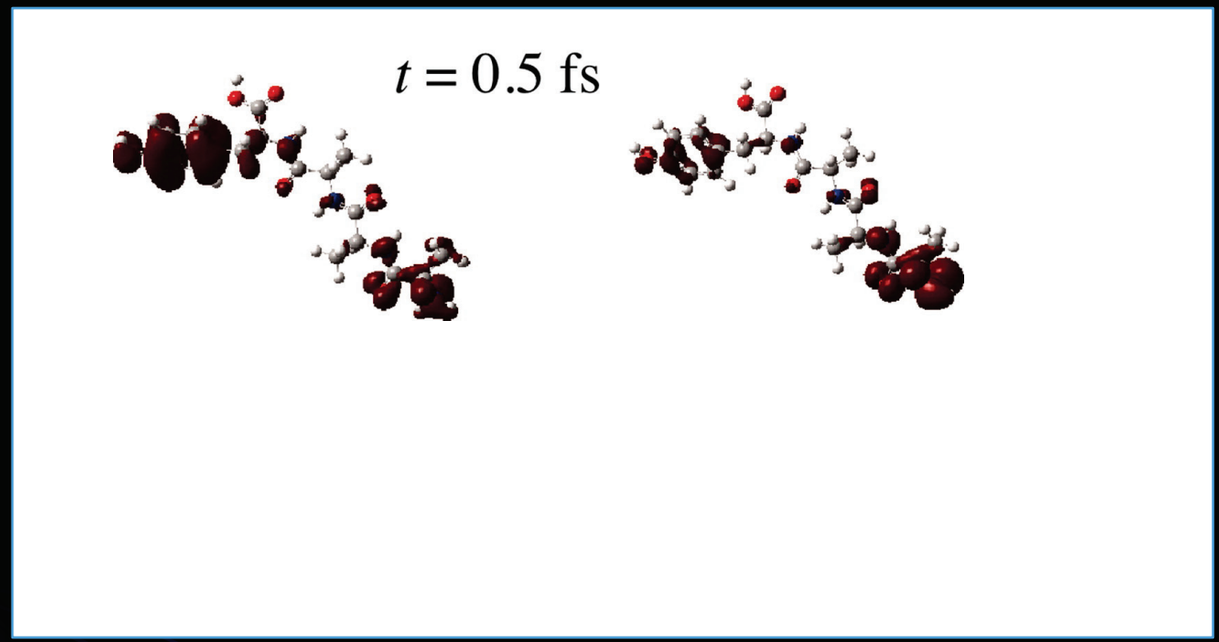

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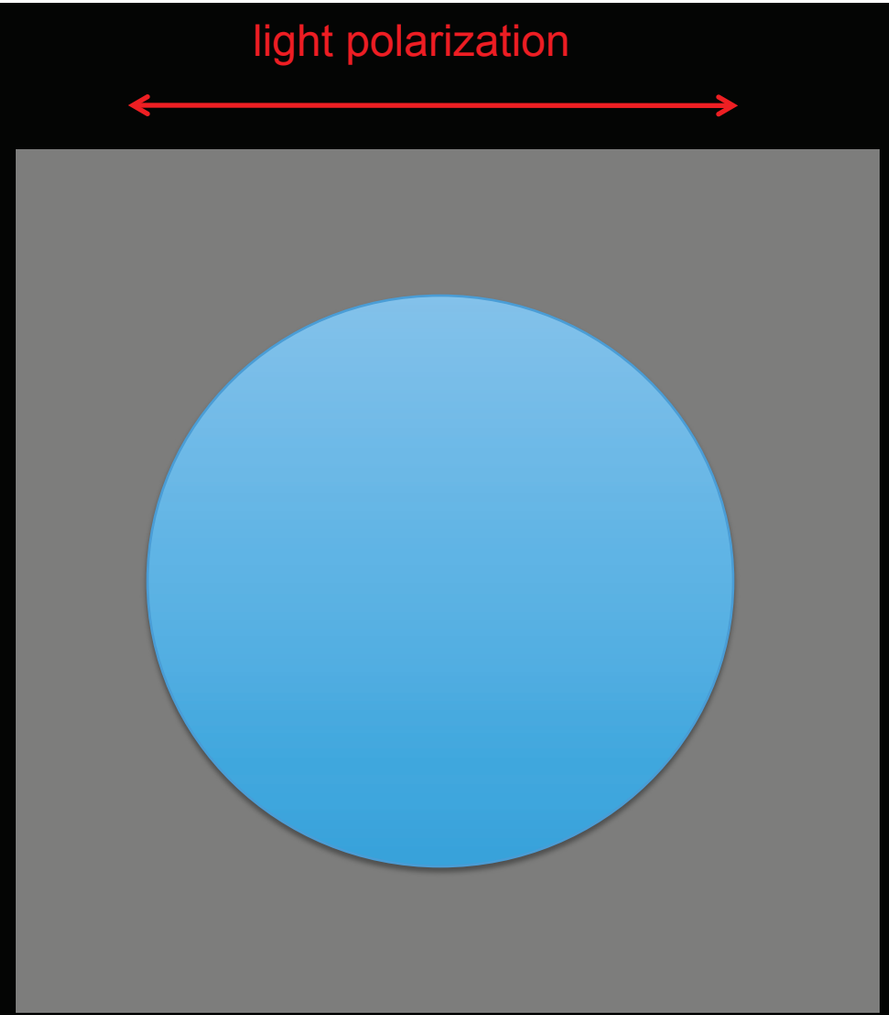
=> Propagation driven by electron correlation

Plasmonic in “real time”

Photoexcited plasmon of a small nanoparticle (1 nm)

TDDFT calculations by

Lauri Lehtovaara
& Miguel Marques



Time-dependant variation of the electron density

Challenges for TD-DFT

Physics with short laser pulses:

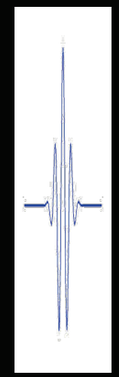
=> strong laser field and XUV photons: ionization and dissociation are common processes in these experiments.

⇒ Challenge for theory (TDDFT) :

- calculate electron, ion spectra
- description of the continuum near the threshold or far above
- description of the long distance interaction between nuclei
- interplay between electronic / nuclear motion
- role of the dissipation
- limitation of the “mean field” approach
- etc …

Challenges for experiments

How can we measure this dynamics?



- light pulses with “relevant properties” (duration, λ)

+

- Relevant “observable” (absorption, photoelectron etc …)

Challenges for experiments

How can we measure this dynamics?

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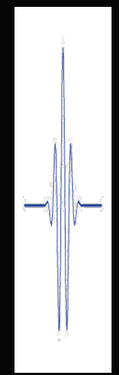
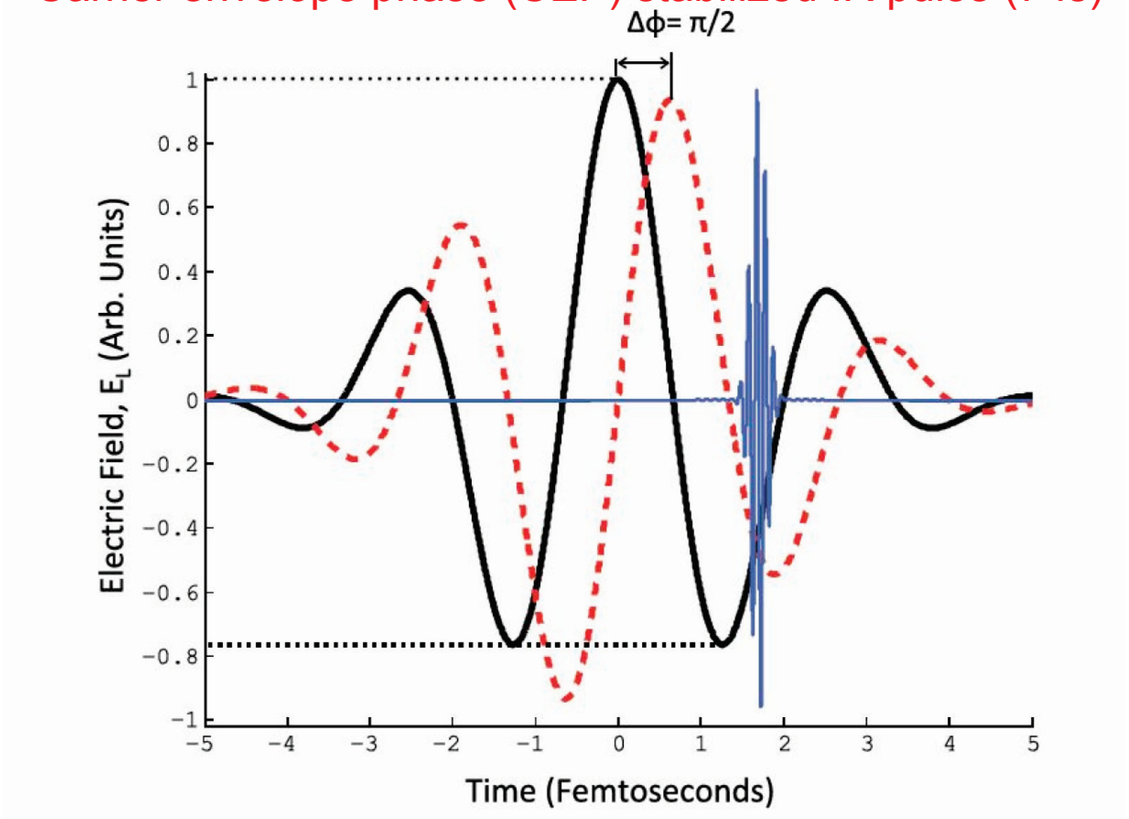


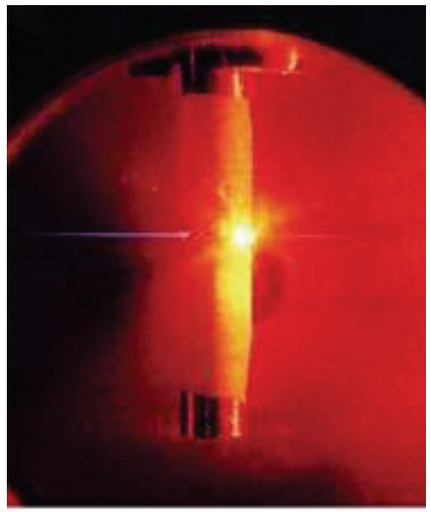
“New” light pulses

Carrier envelope phase (CEP) stabilized IR pulse (7 fs)



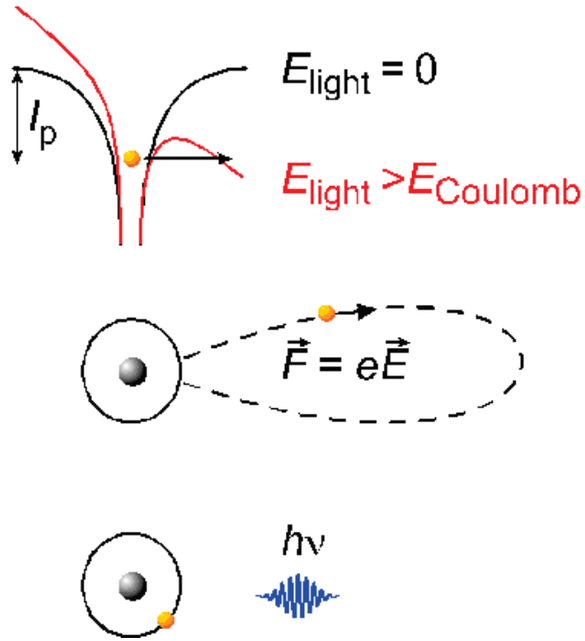
From strong laser laser field…

to synthesis of attosecond pulses



The Synthesis of attosecond pulses is based on a nonlinear, non-perturbative process: High Harmonic Generation that occurs when an intense laser beam is focussed on a gas sample.

This process is explained by the so-called 3 steps model:



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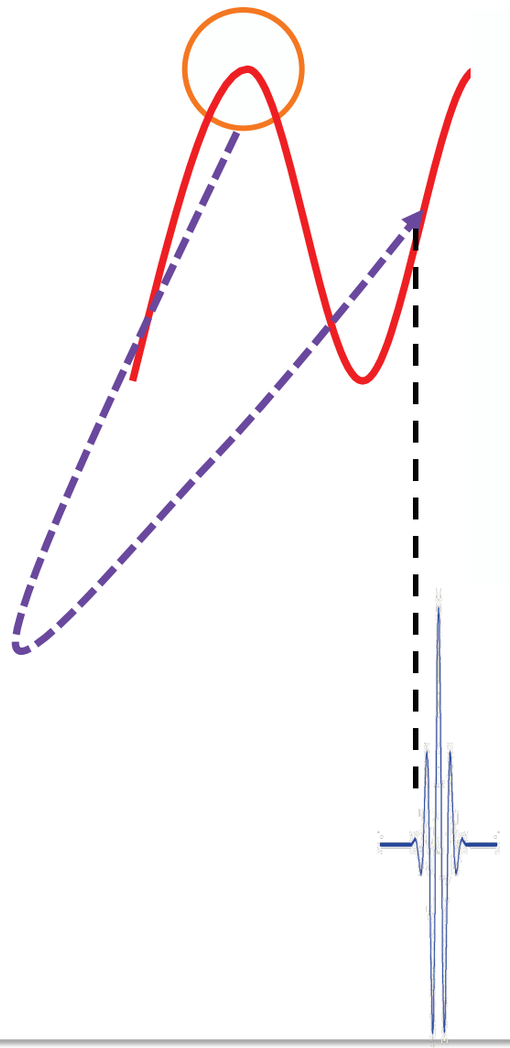
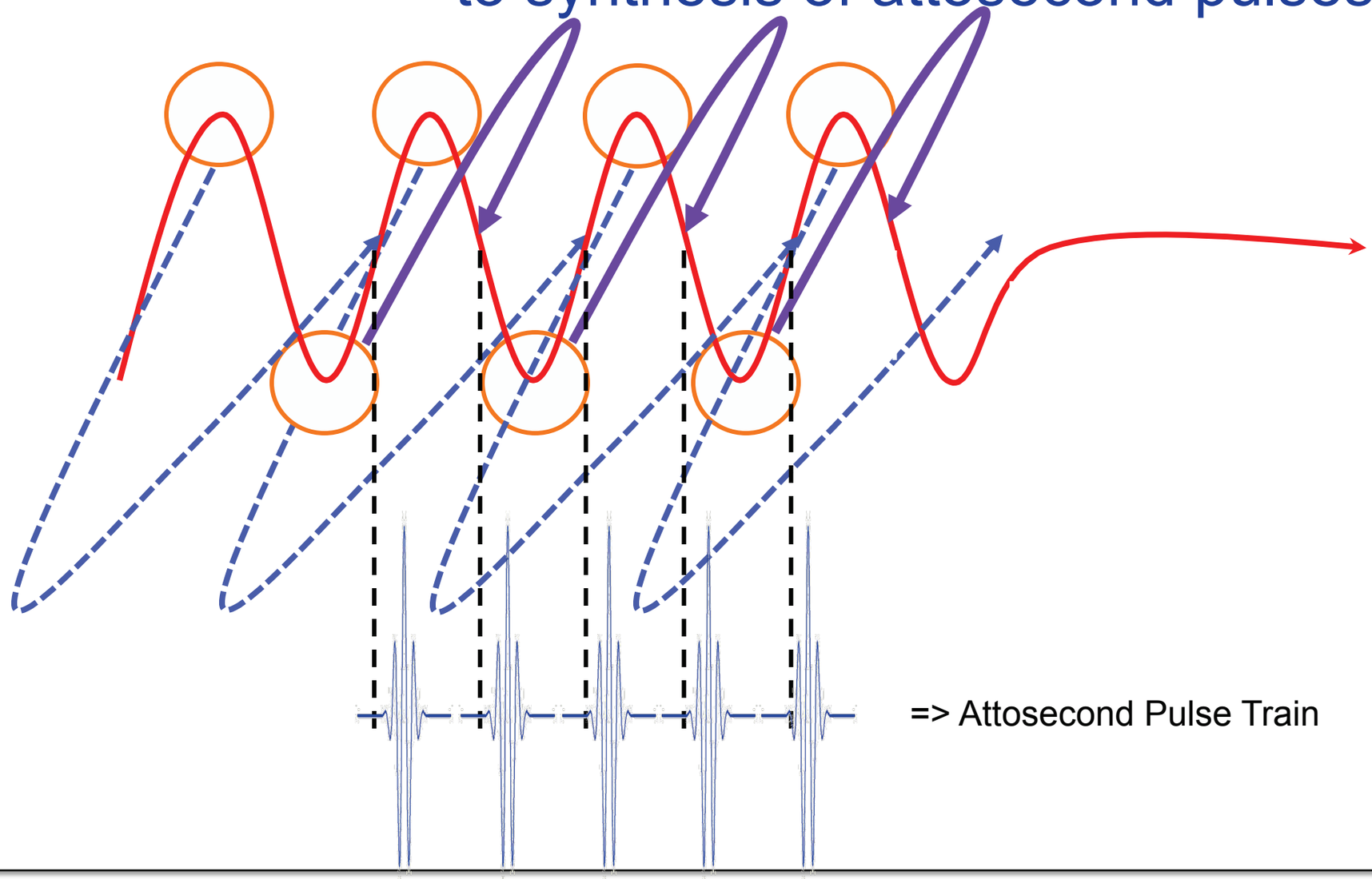


=> Single Attosecond Pulse

From strong laser laser field…

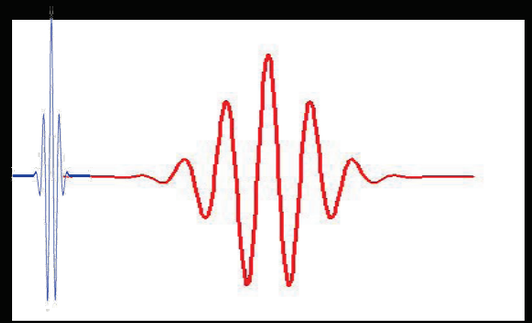
to synthesis of attosecond pulses

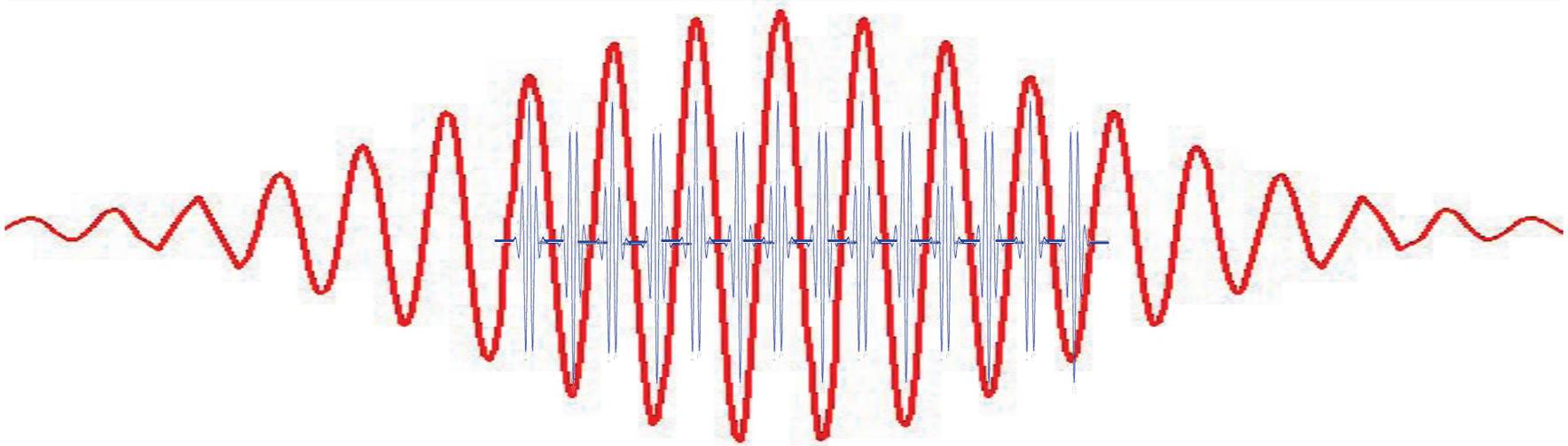


“New” light pulses

Sub-femtosecond electron dynamics using a pump-probe set-up:

Short IR phase locked pulse in combination with attosecond pulse (APT or Single).

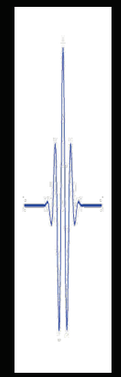




The delay between the pulses can be controlled (=> very high stability)

Challenges for experiments

How can we measure this dynamics?

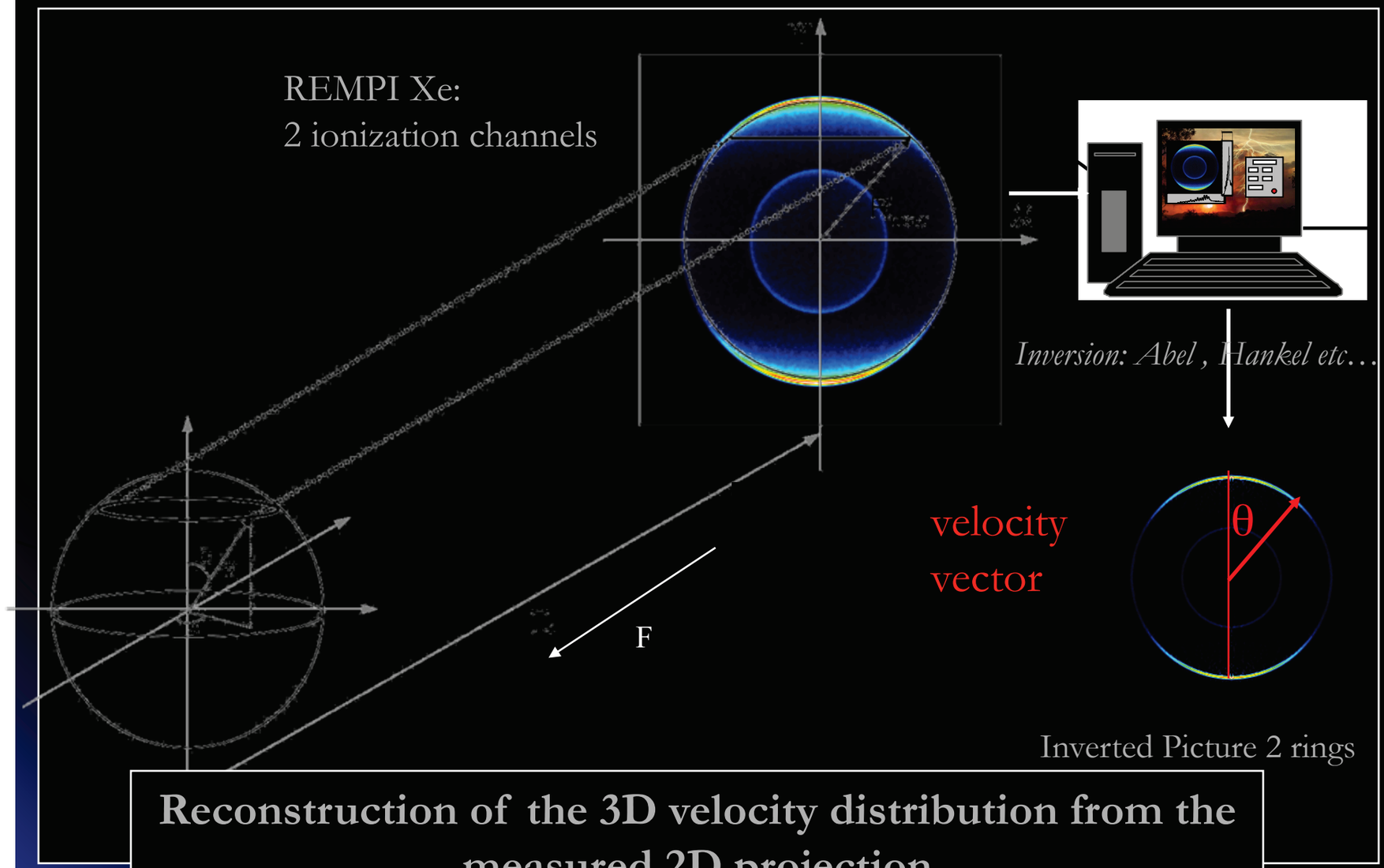


- light pulses with “relevant properties” (duration, λ)

+

- Relevant “observable” (absorption, photoelectron etc …)

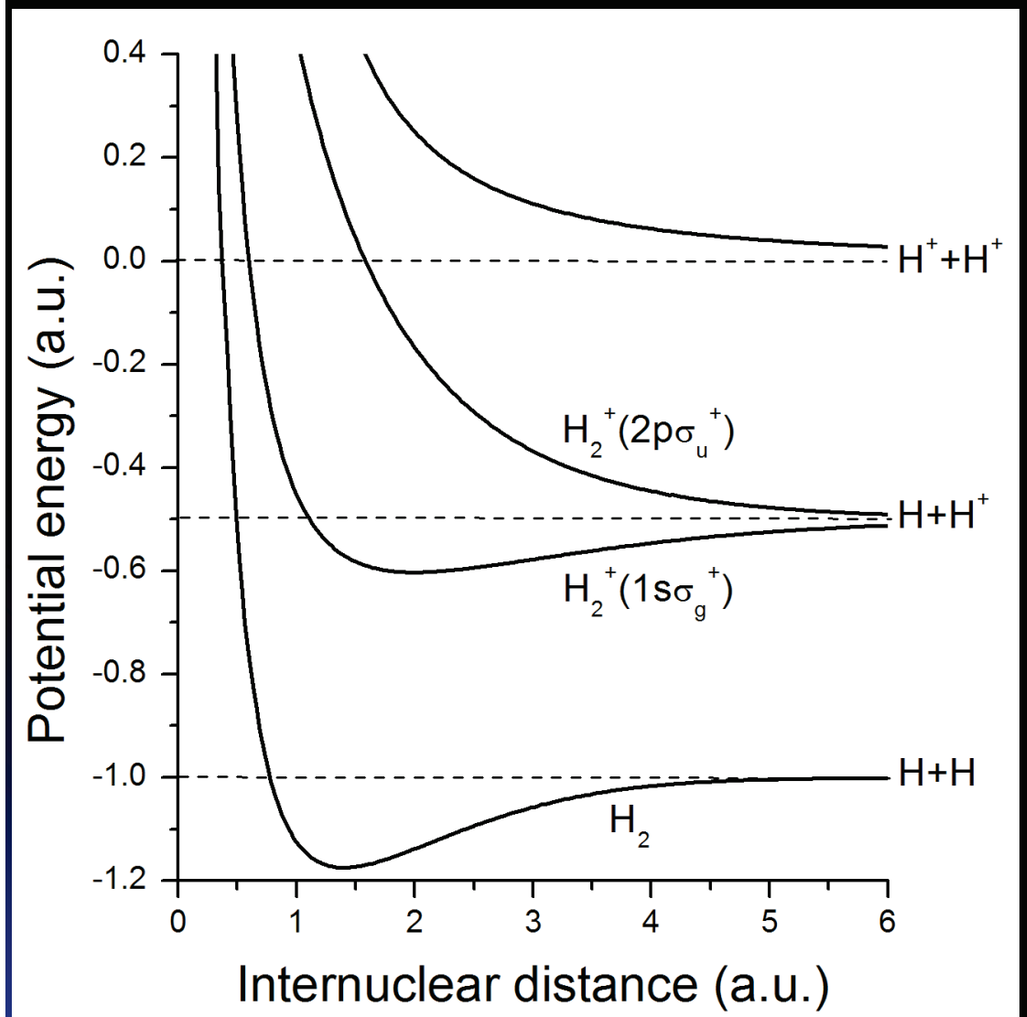
Observable : electron/ion momentum distribution



Reconstruction of the 3D velocity distribution from the measured 2D projection

-II- An example of attosecond experiments: Attosecond electron localization spectroscopy

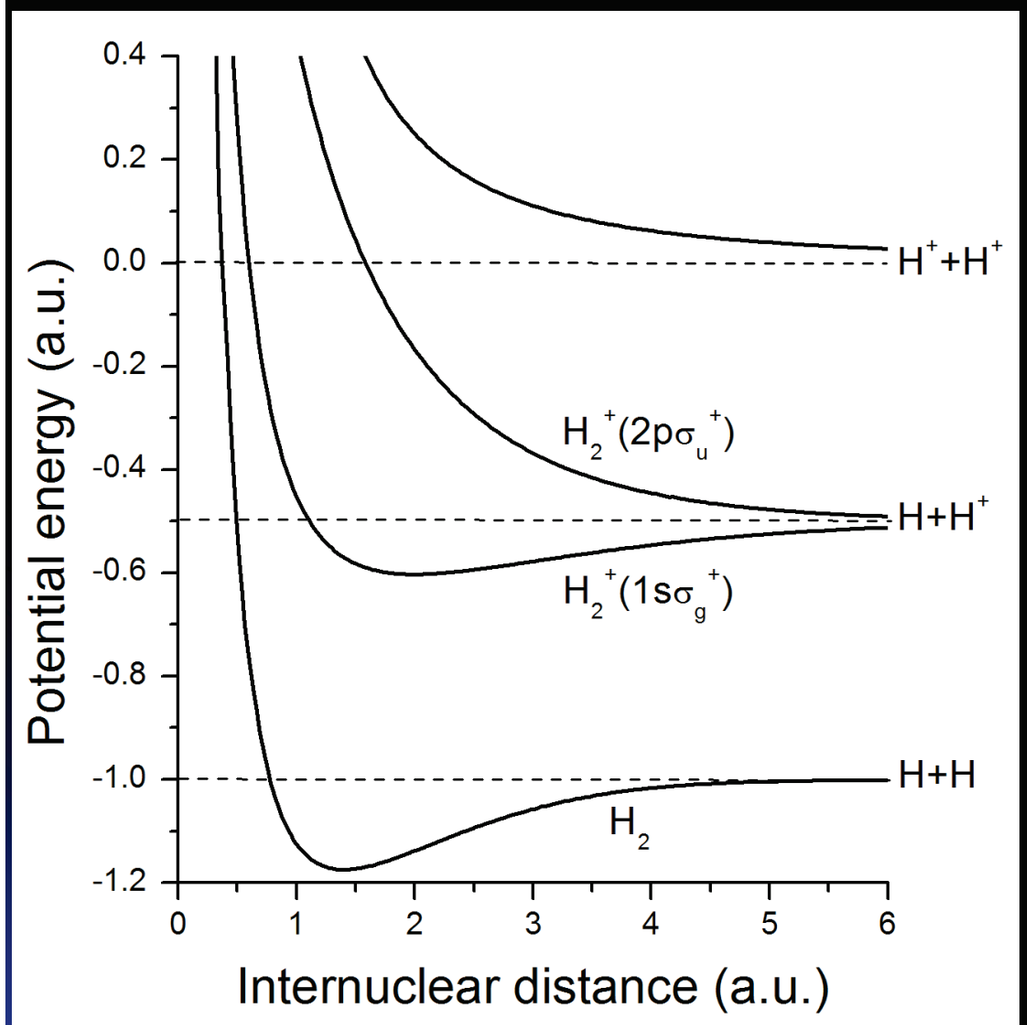
-II- Attosecond electron localization spectroscopy



dissociative ionization

$H_2 + hv \rightarrow H + H^+ + e$

-II- Attosecond electron localization spectroscopy



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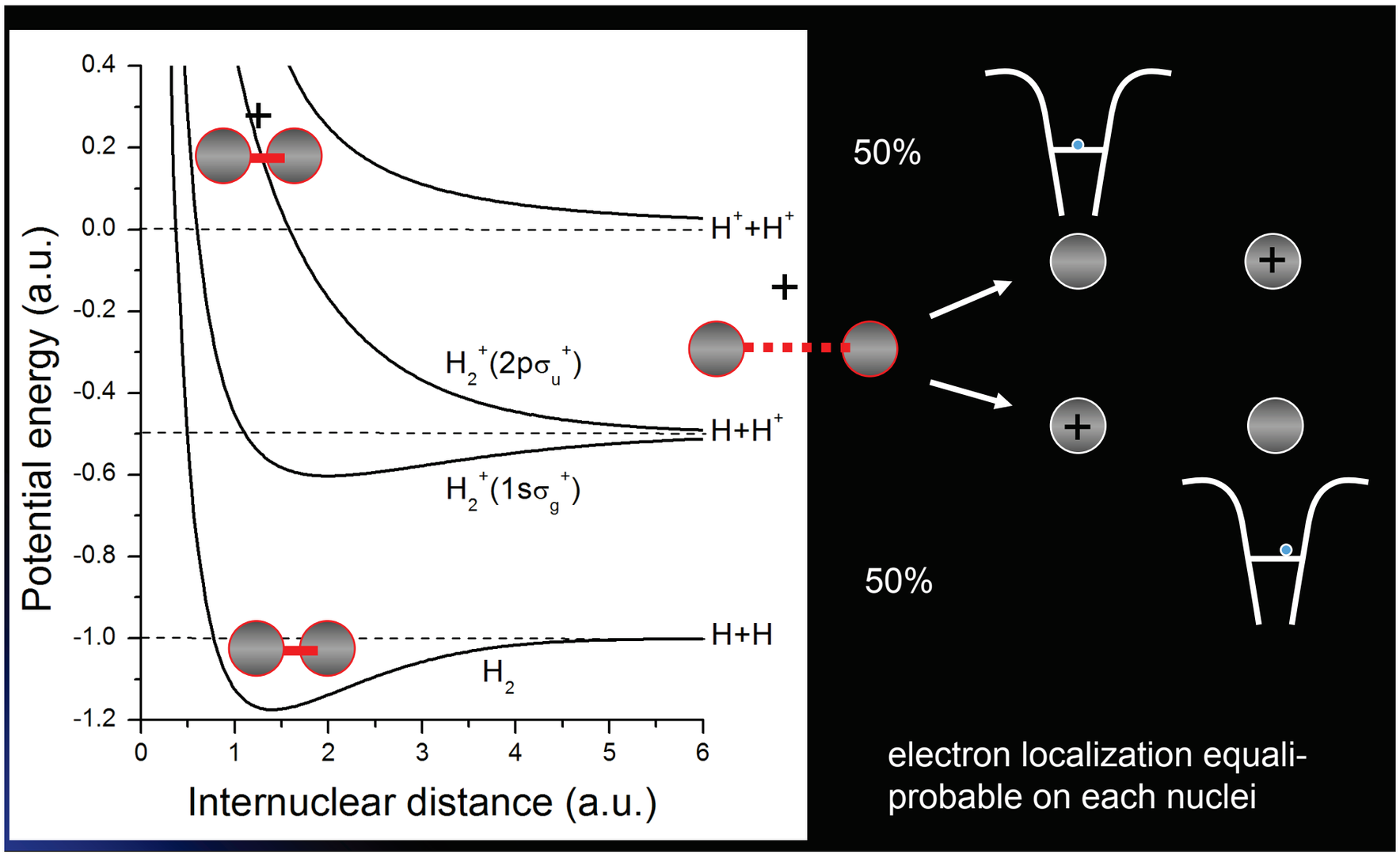
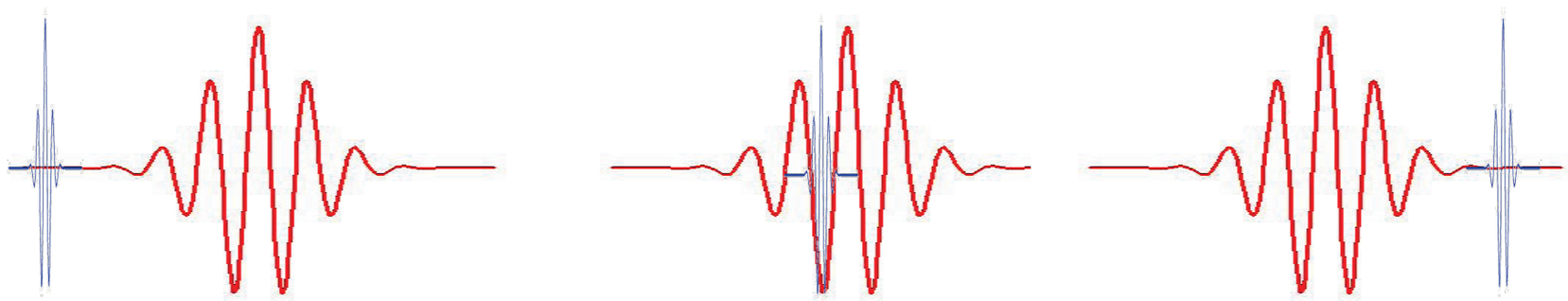


-II- Attosecond electron localization spectroscopy

Can we control the electron localization on the attosecond timescale using short pulses?



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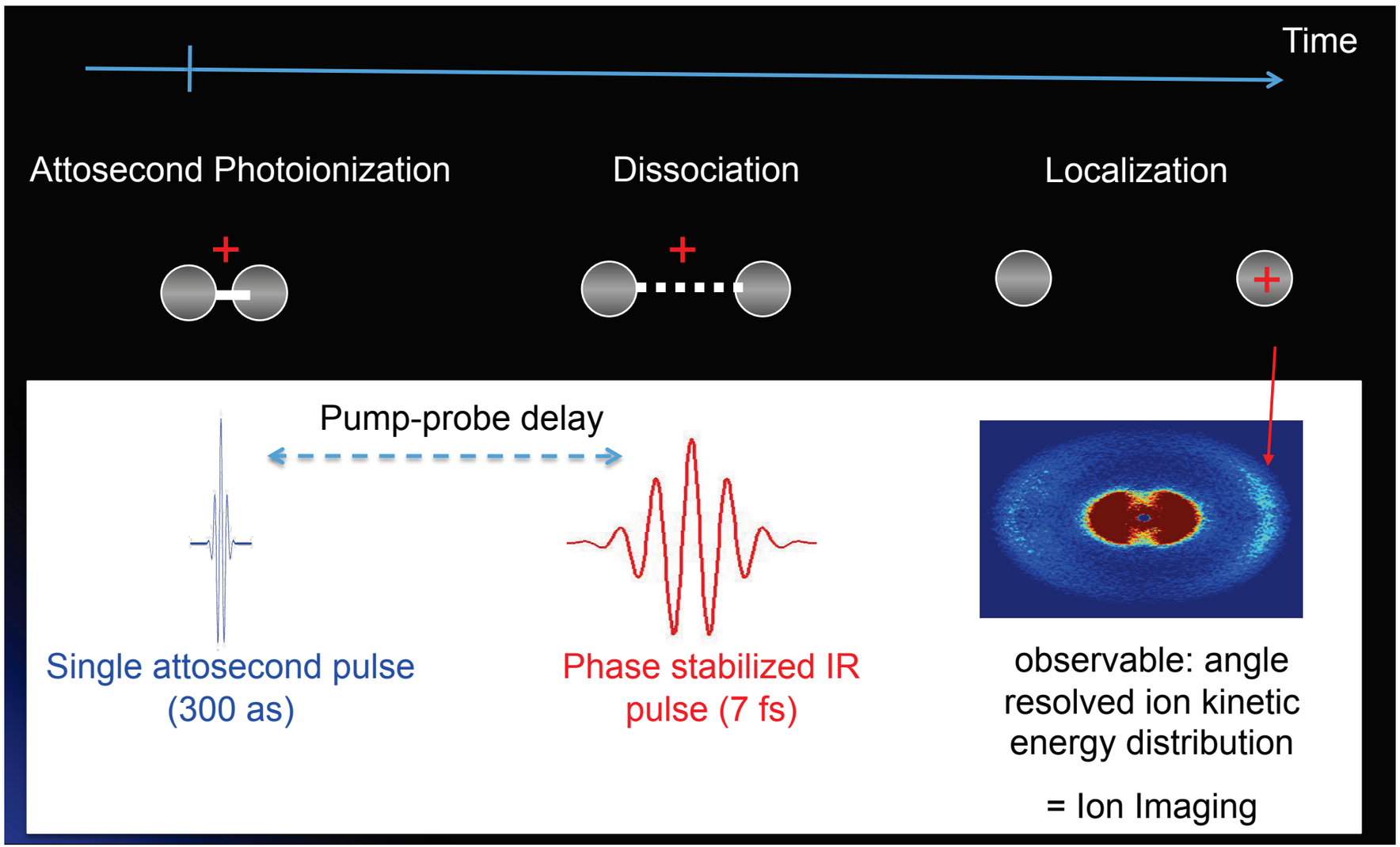

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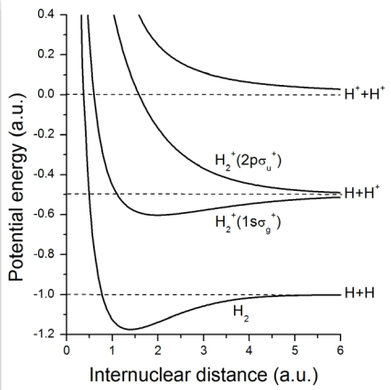

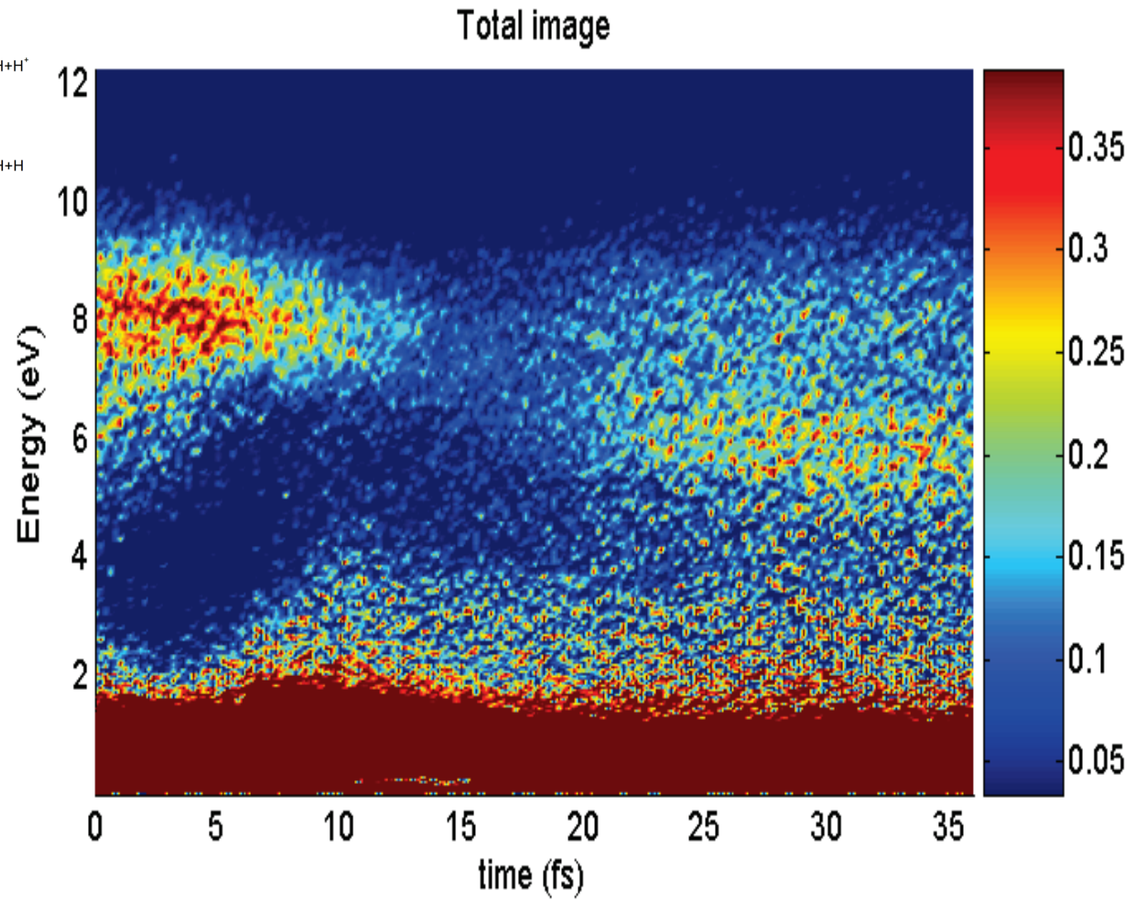

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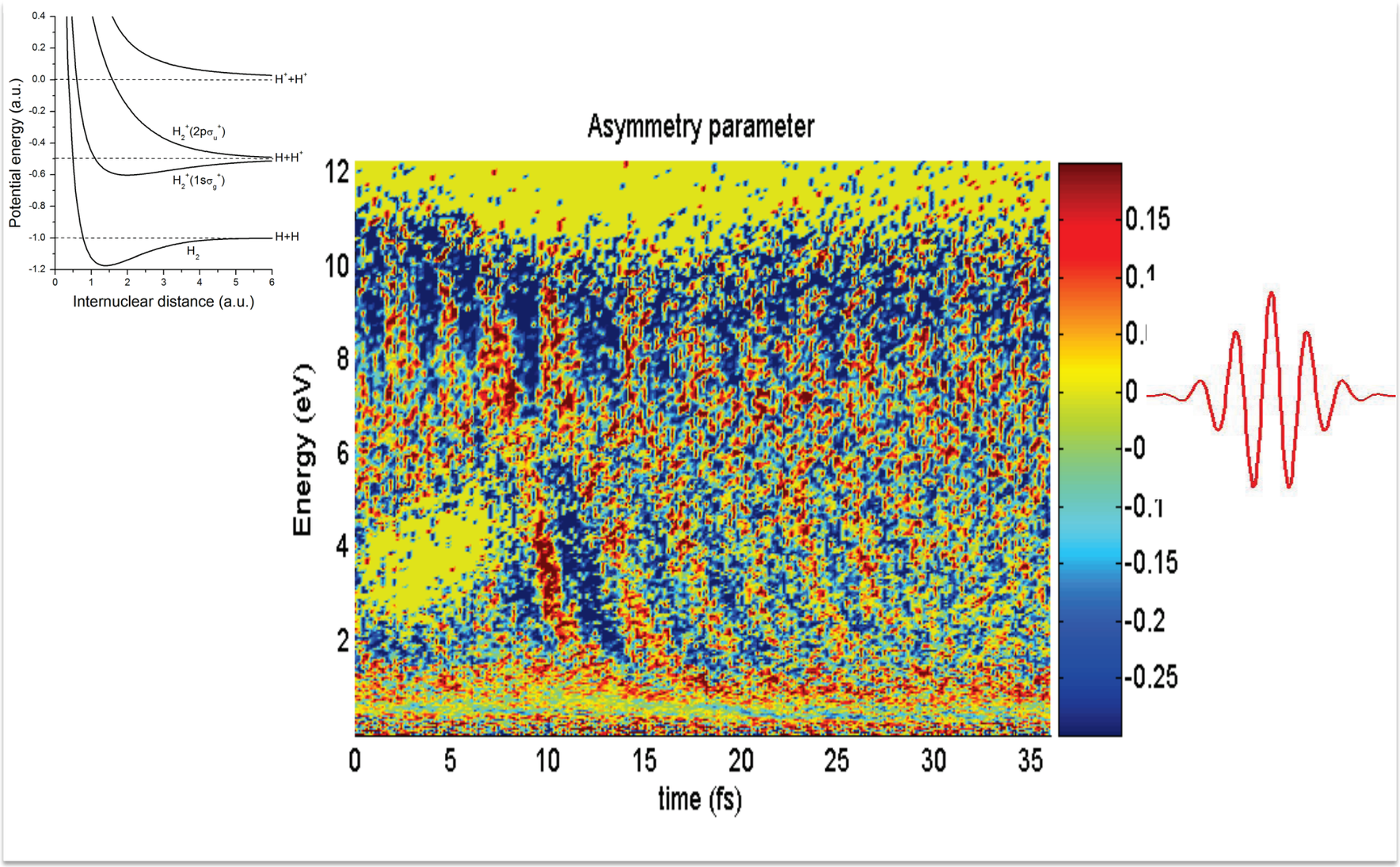

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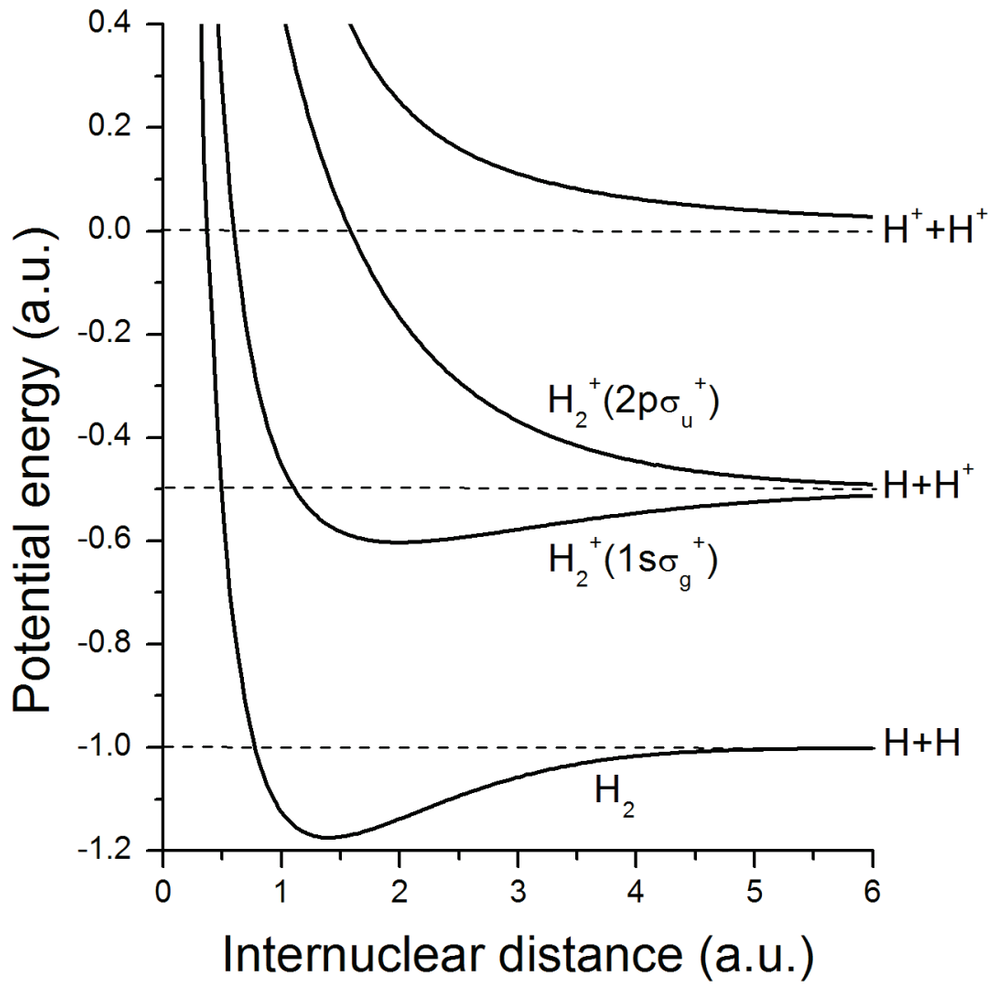

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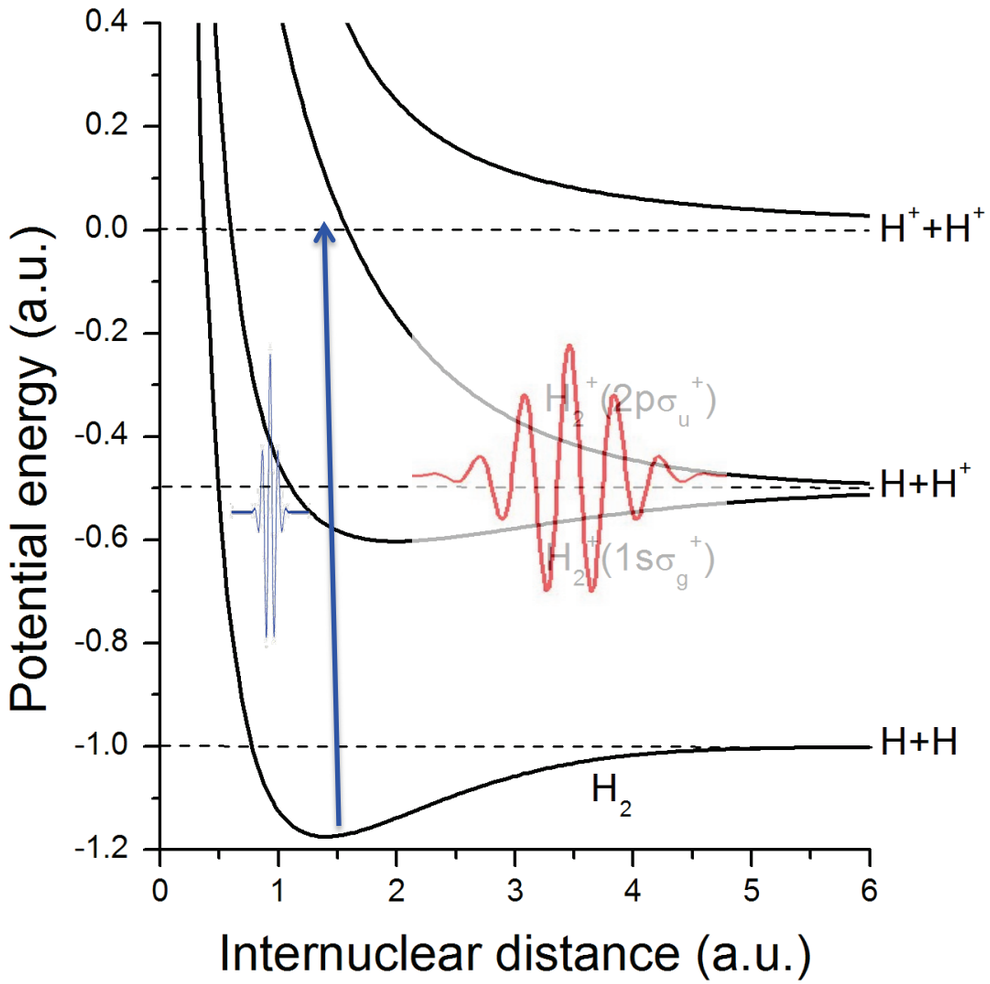
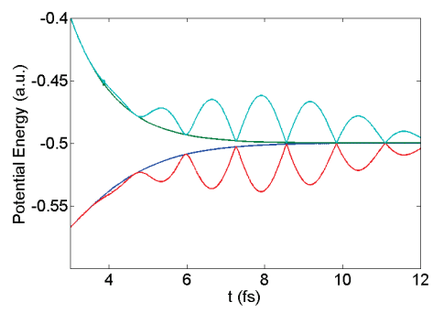
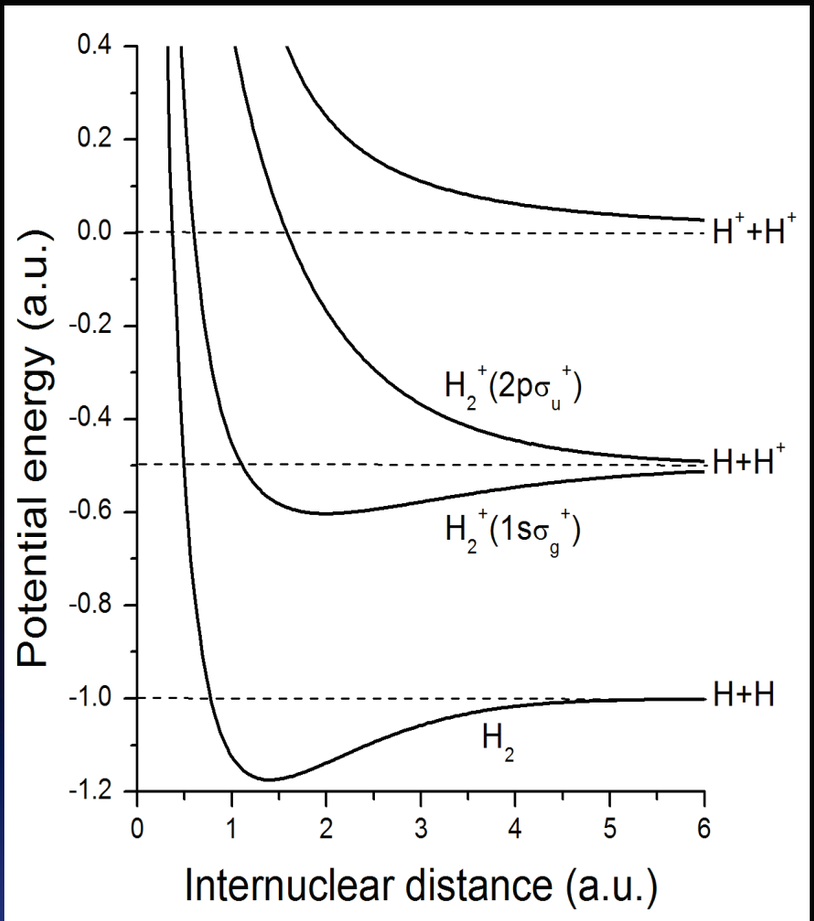


"Landau-Zener" coupling due to the IR field determines the final wavefunction as a coherent superposition of the σ_u and σ_g state

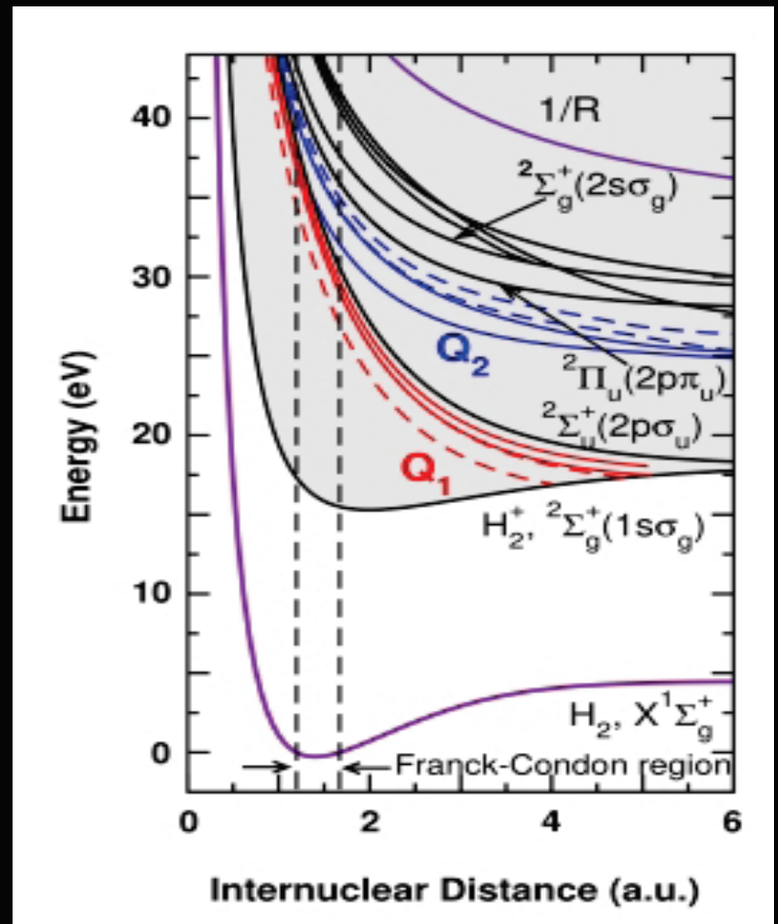


-II- Attosecond electron localization spectroscopy

Single active electron picture



+ Doubly excited states



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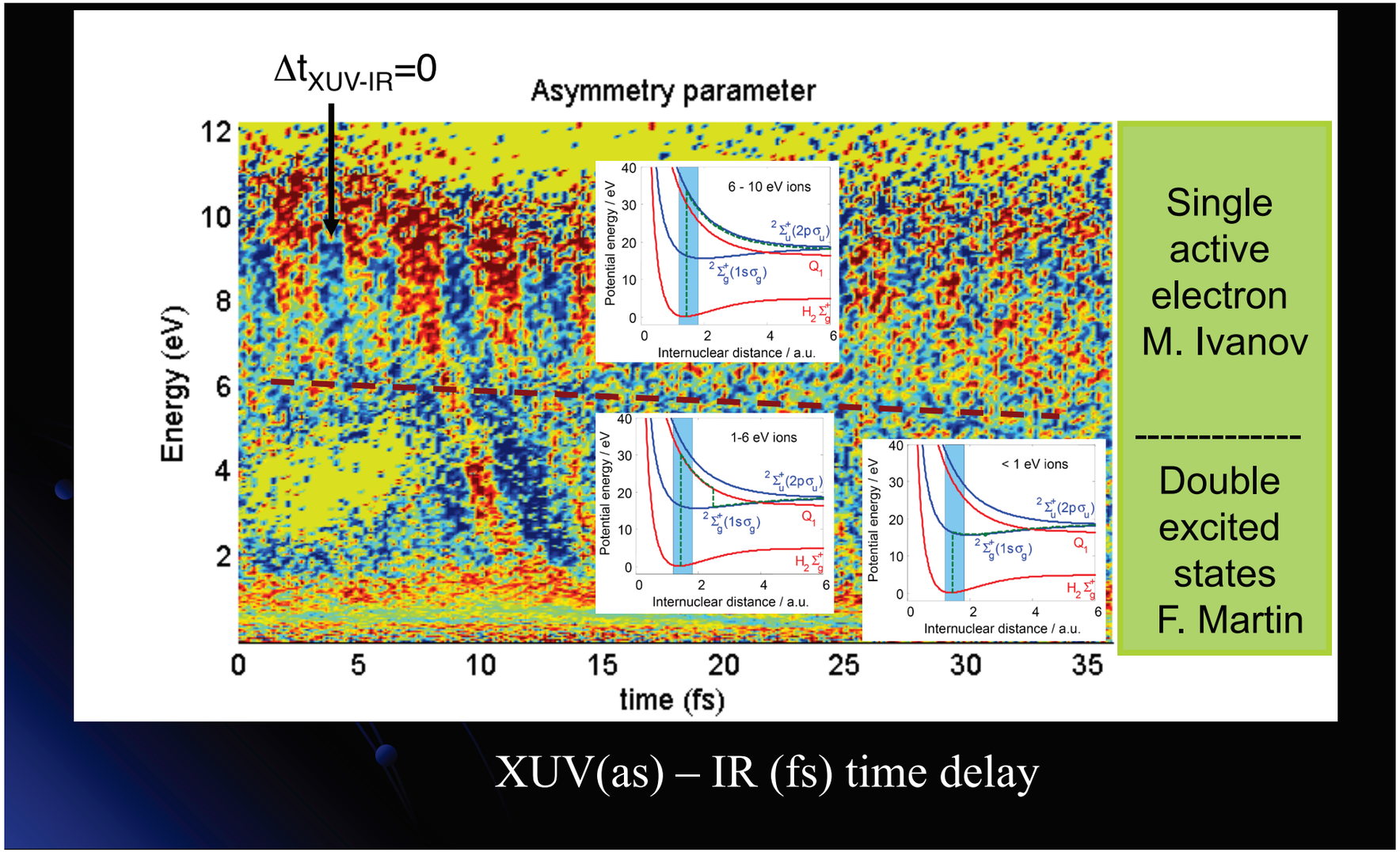
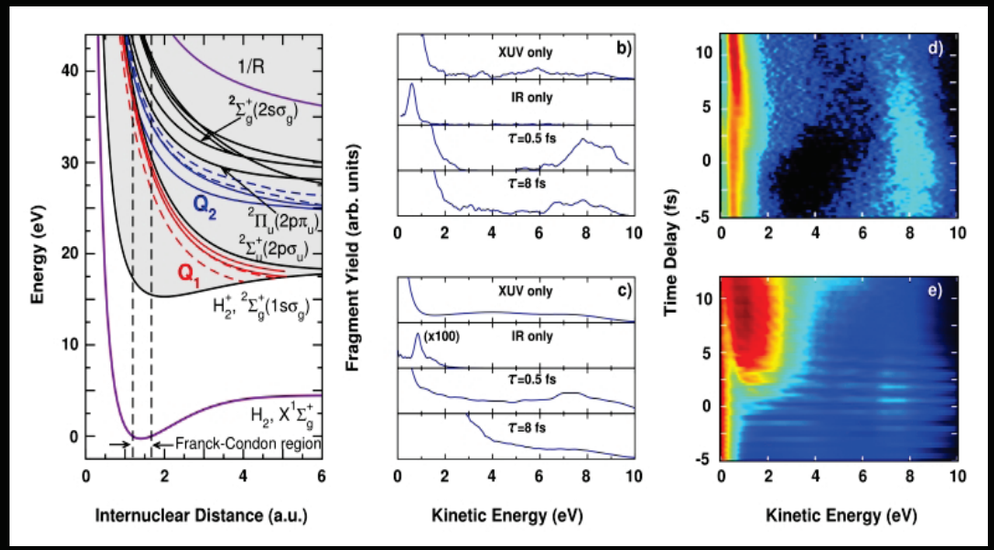


XUV(as) – IR (fs) time delay

-II- Attosecond electron localization spectroscopy



⇒ Creation of an electro-nuclear attosecond wavepacket

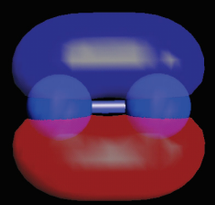
⇒ Manipulation of the molecular potential energy surfaces with light : dressed states / L.Z coupling

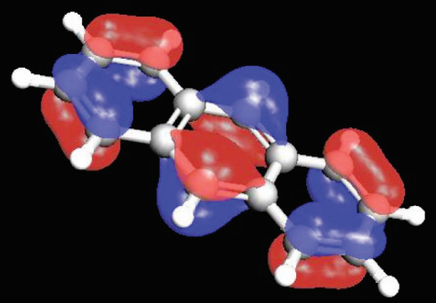
⇒ Control of the final photo-product:

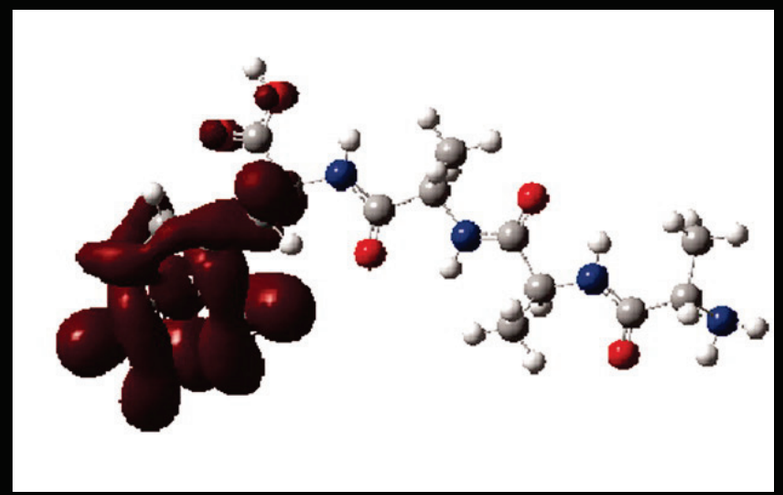
=> Photo-chemistry on the electronic timescale

-II- Attosecond electron localization spectroscopy

Photo-chemistry on the electronic timescale







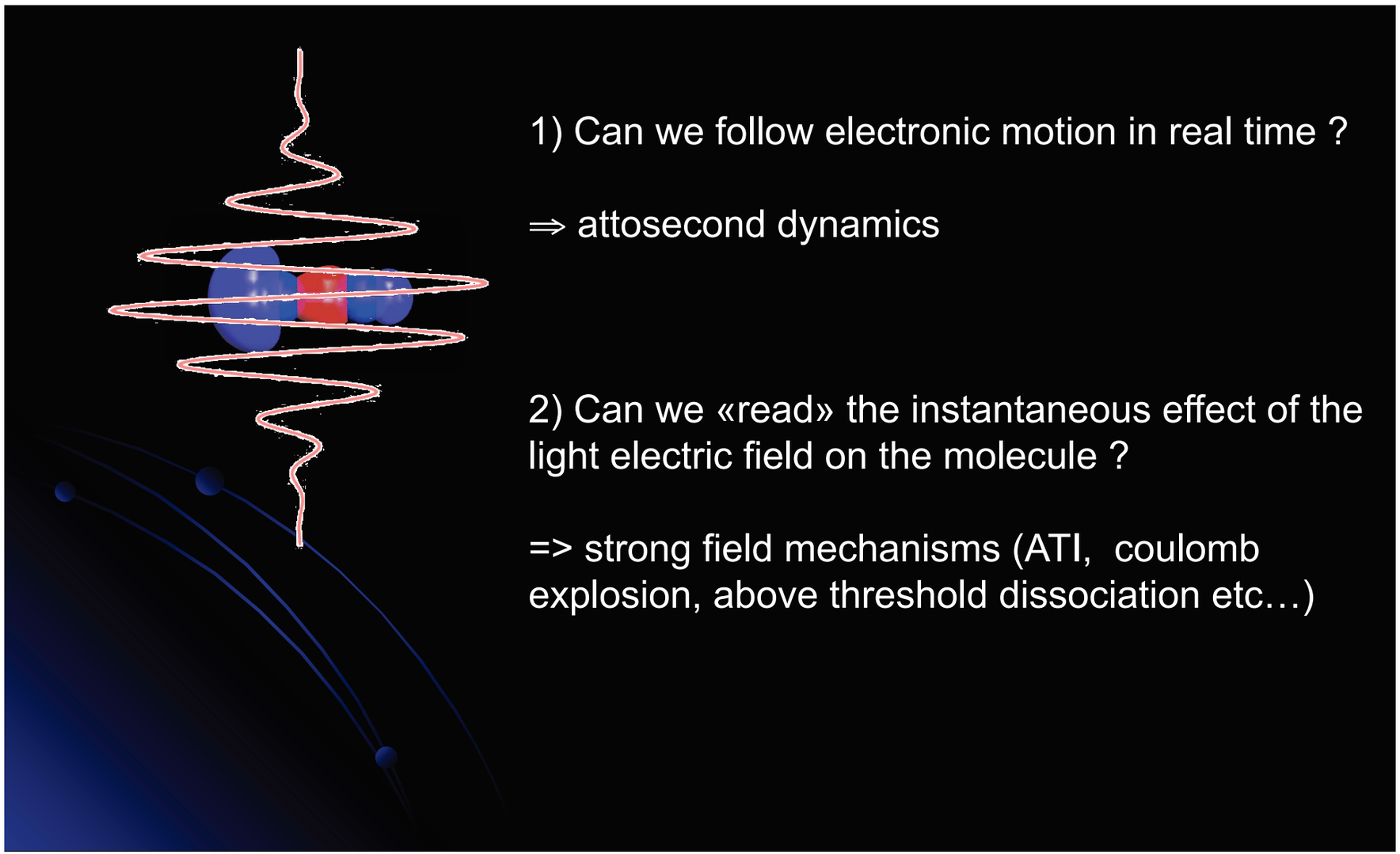
larger molecules

role of the - coherence?
- nuclear motion?
- various quantum states
- dissipation, stochastic mechanisms?

real time / real space
TD-DFT ?

-III- other attosecond experiments with molecules …

Transient attosecond dynamics



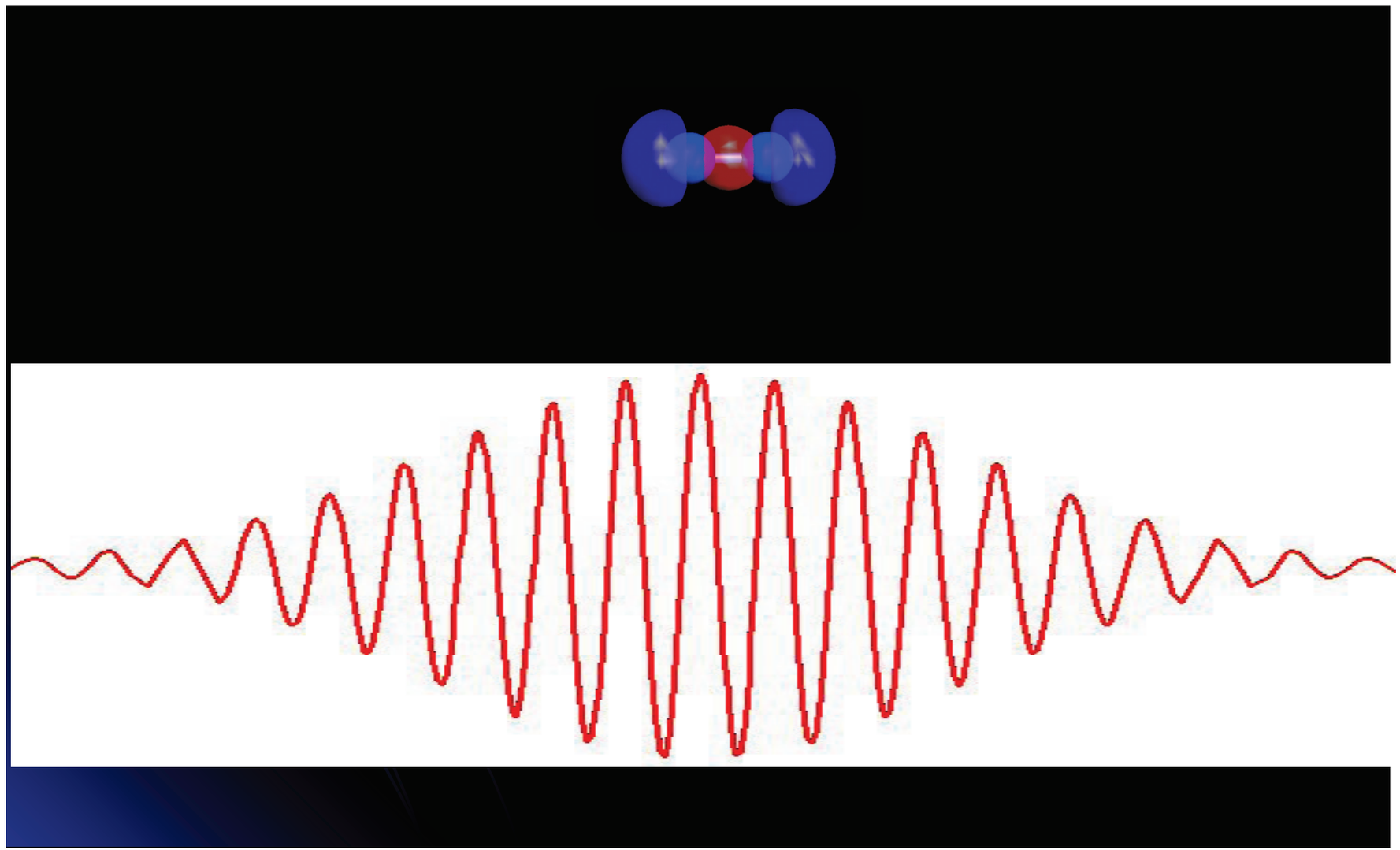
1) Can we follow electronic motion in real time ?

⇒ attosecond dynamics

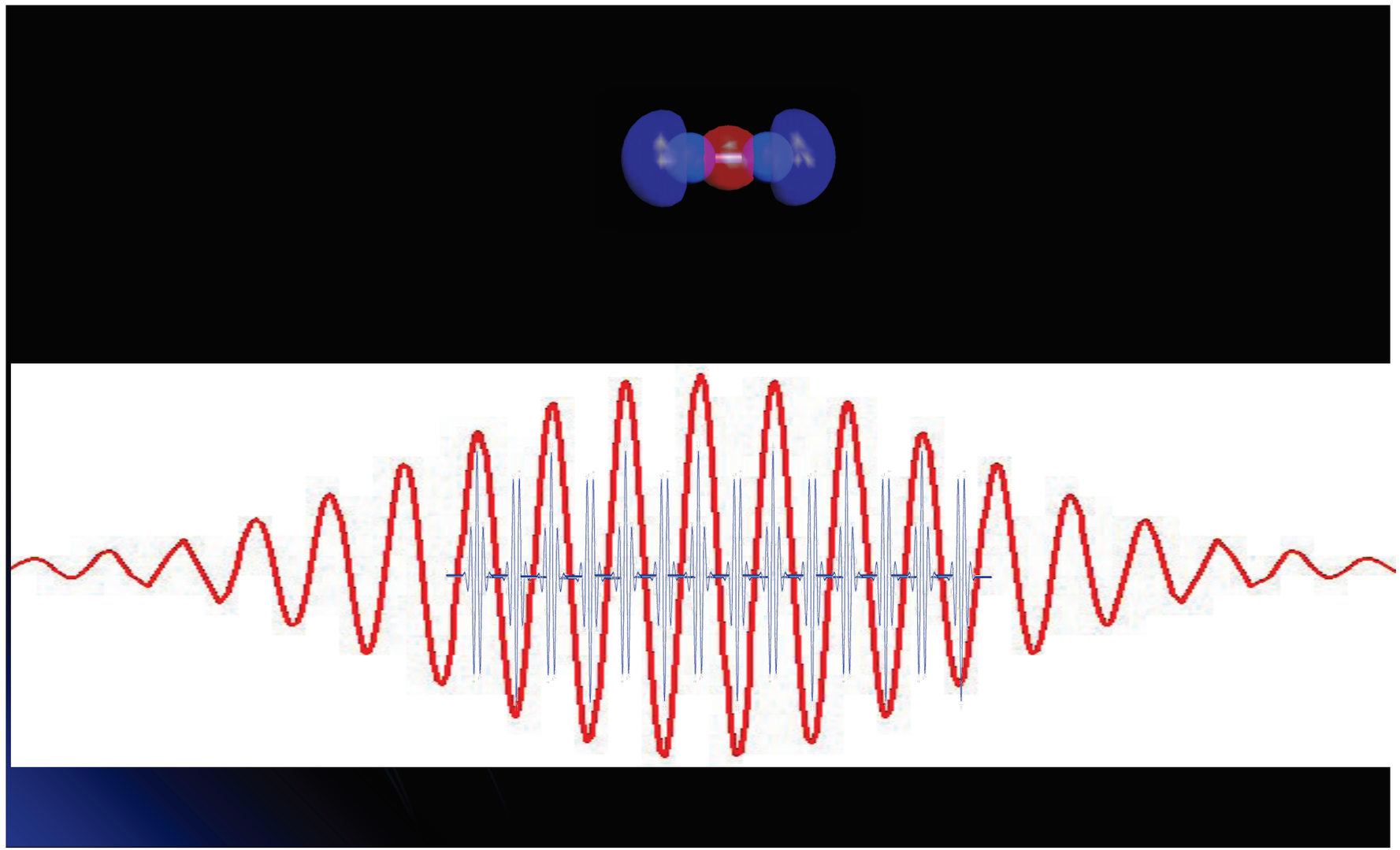
2) Can we «read» the instantaneous effect of the light electric field on the molecule ?

=> strong field mechanisms (ATI, coulomb explosion, above threshold dissociation etc…)

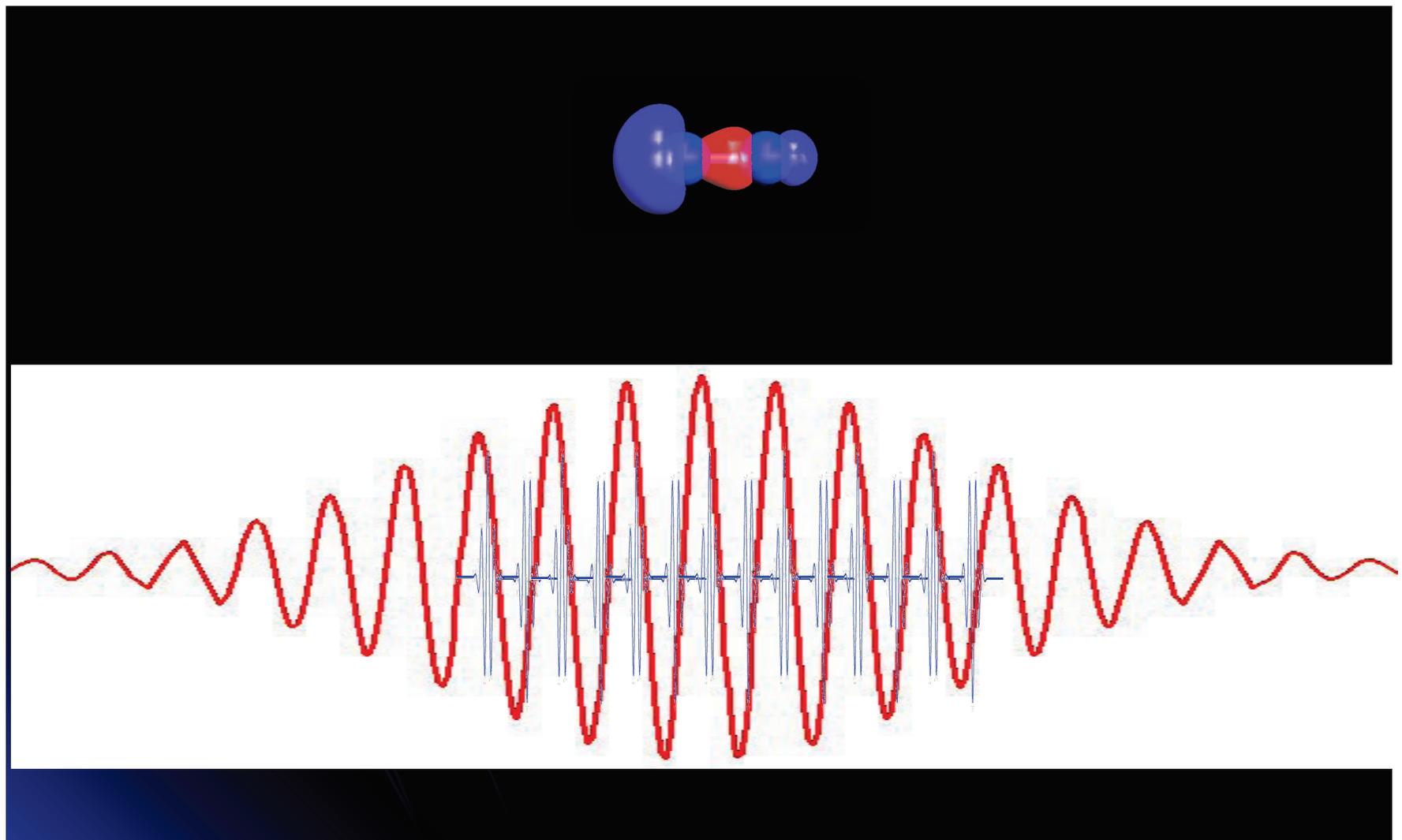
Transient attosecond dynamics



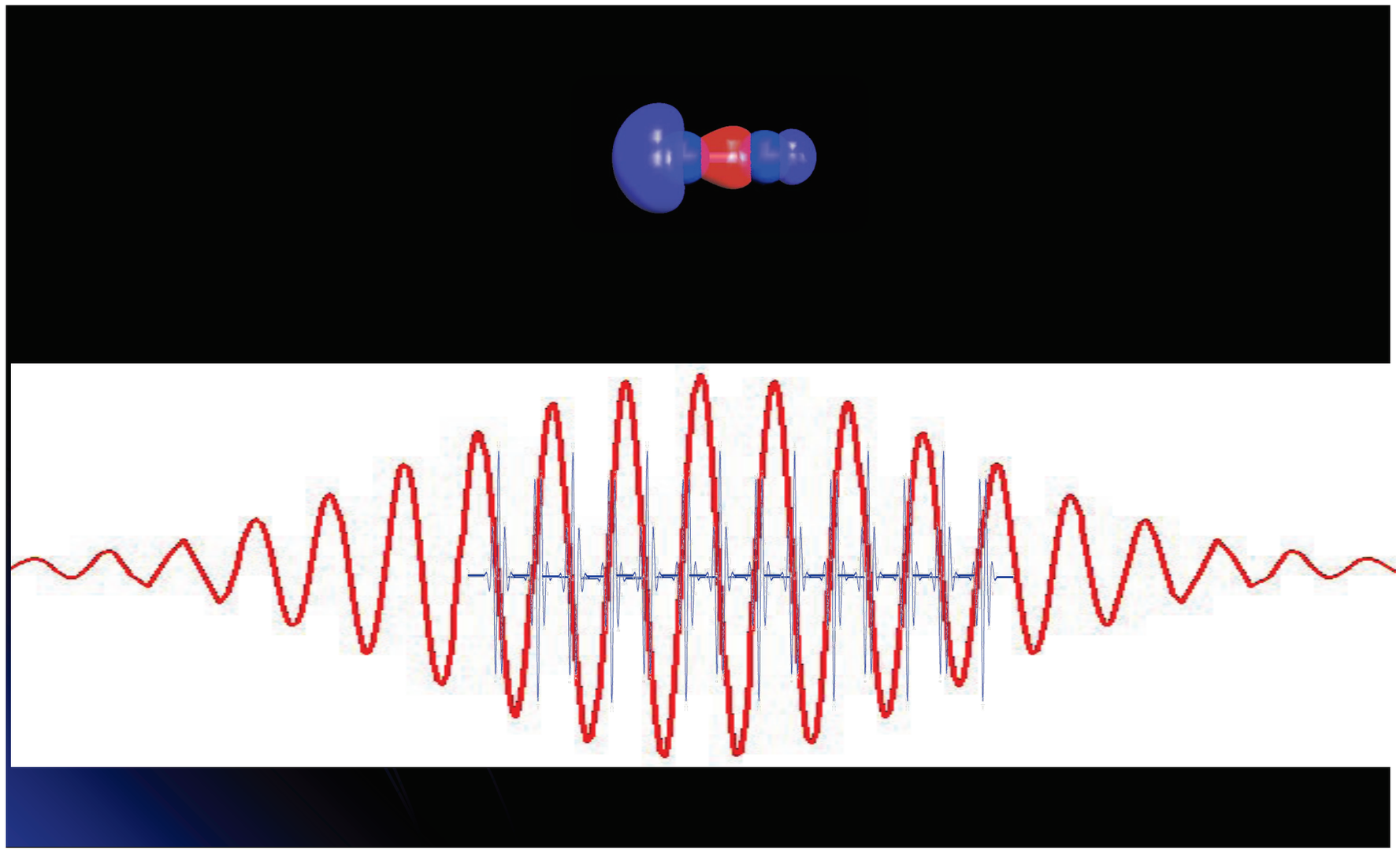
Transient attosecond dynamics



Transient attosecond dynamics



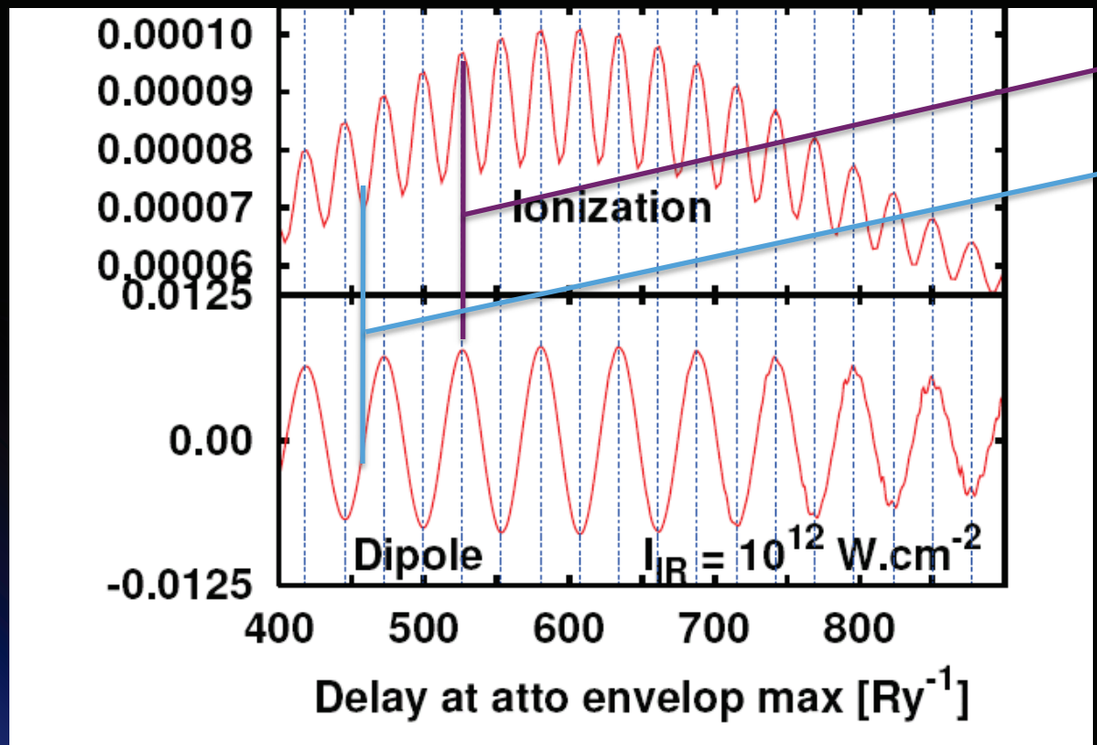
Transient attosecond dynamics

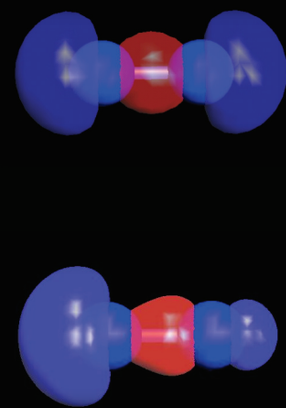


Transient attosecond dynamics

Molecular photoionization:

N_2 + XUV pulse -> N_2^+ + e





TD-DFT calculations
Coll. Group of E. Suraud

Transient attosecond dynamics

- Photoionization

$$N_2 \rightarrow N_2^+ + e$$

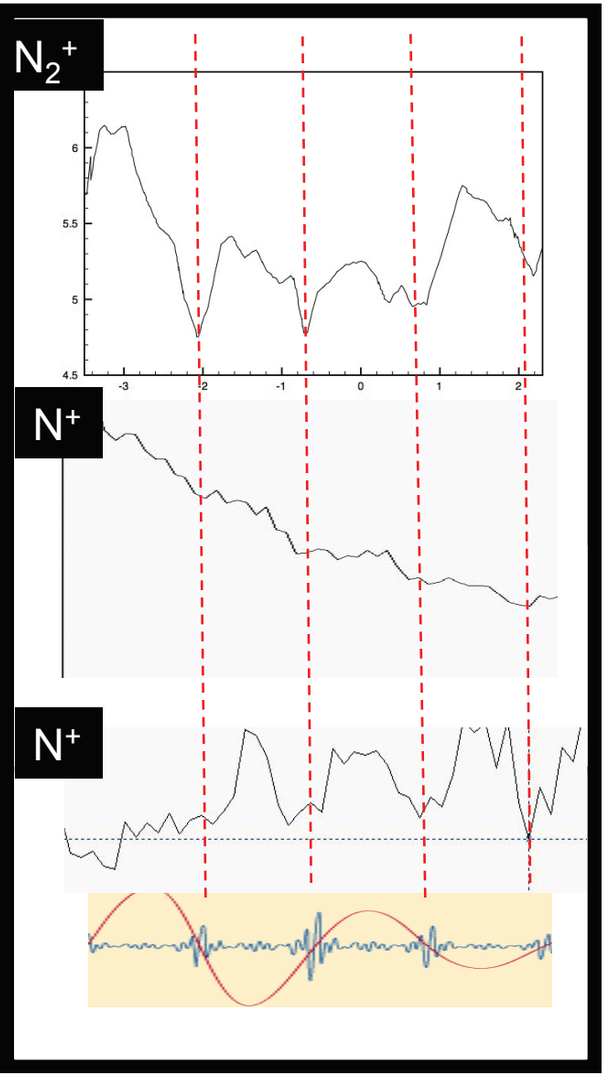
- Dissociative ionization

$$N_2 \rightarrow N^+ + N + e$$

- Coulomb explosion

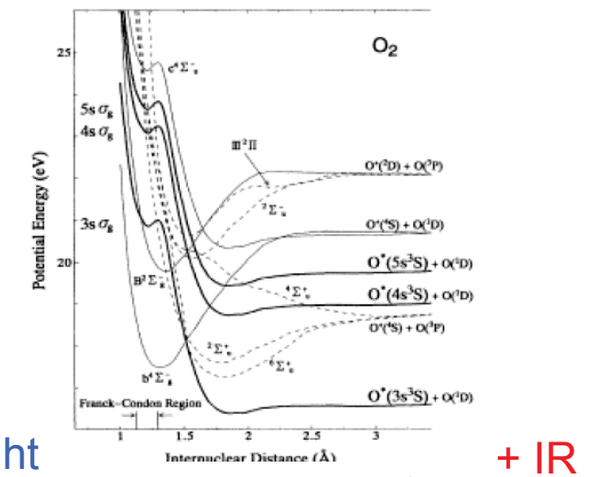
$$N_2 \rightarrow N^+ + N^+ + 2e$$

Experiment performed in Lund
(coll. group of Anne L'Huillier)

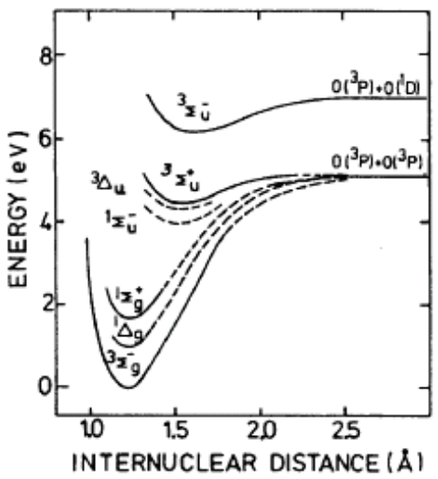


Transient attosecond dynamics

XUV light



+ IR



IR dressed molecule

-

XUV excitation

-

Leads to Ionization, dissociation etc…

-> Measure of velocity distribution of the particles

-> try to « read » the dressed properties of the molecule

Transient attosecond dynamics

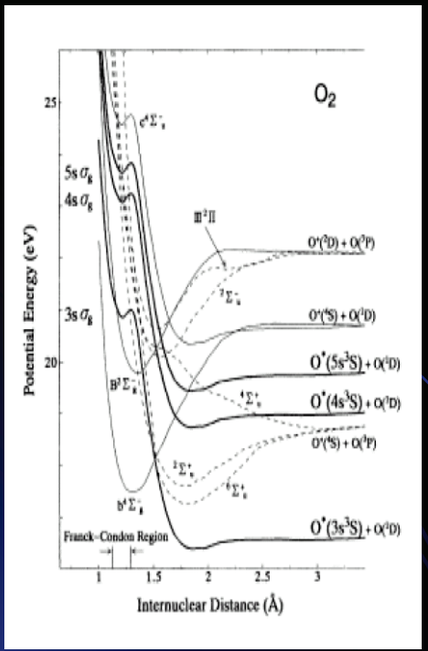
molecular photodissociation

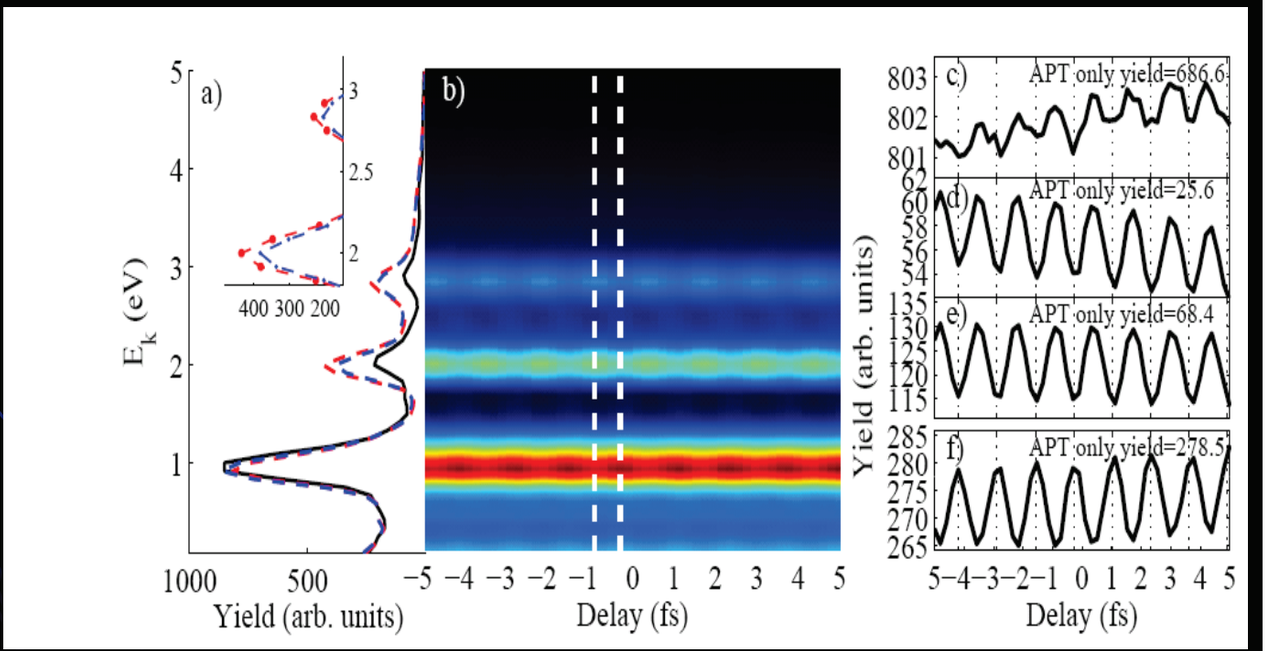
$O_2 \rightarrow O^+ + O + e$

Ion kinetic energy spectrum

experiment performed at Amolf
Group of Marc Vrakking

manuscript in preparation …

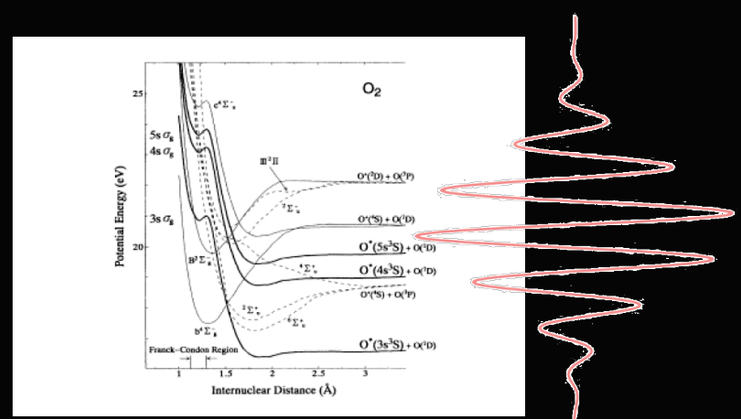




Transient attosecond dynamics

Probe in real-time the electronic motion in a molecule during its interacting with light (transient dynamics)

Coherent control of the ionization, dissociation, coulomb explosion on a sub-cycle level



role of the - light properties?
- resonances ?
- nuclear motion?
- various quantum states ?
- dissipation, stochastic mechanisms?

real time / real space
TD-DFT ?

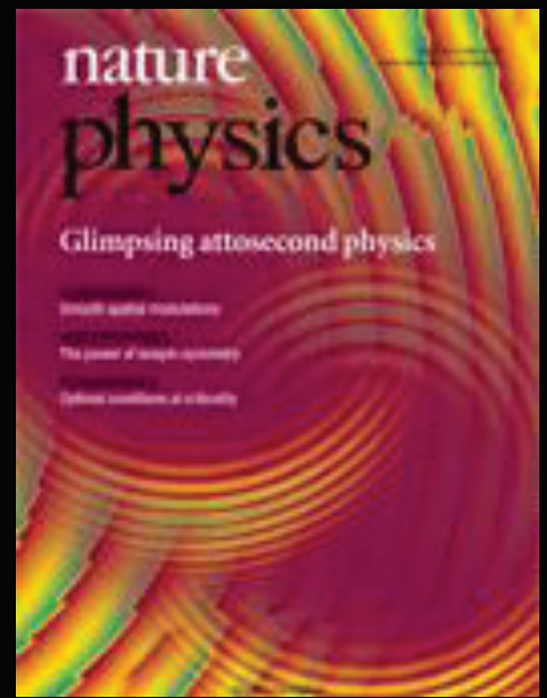
[redacted]

-IV- “Rabbit” & “Streaking”

Streaking

=> Very important process: « electron streaking »

An electron interacting with a strong light field experiences an acceleration by the light electric field. The electron final momentum depends on the integral of the field (potential vector).



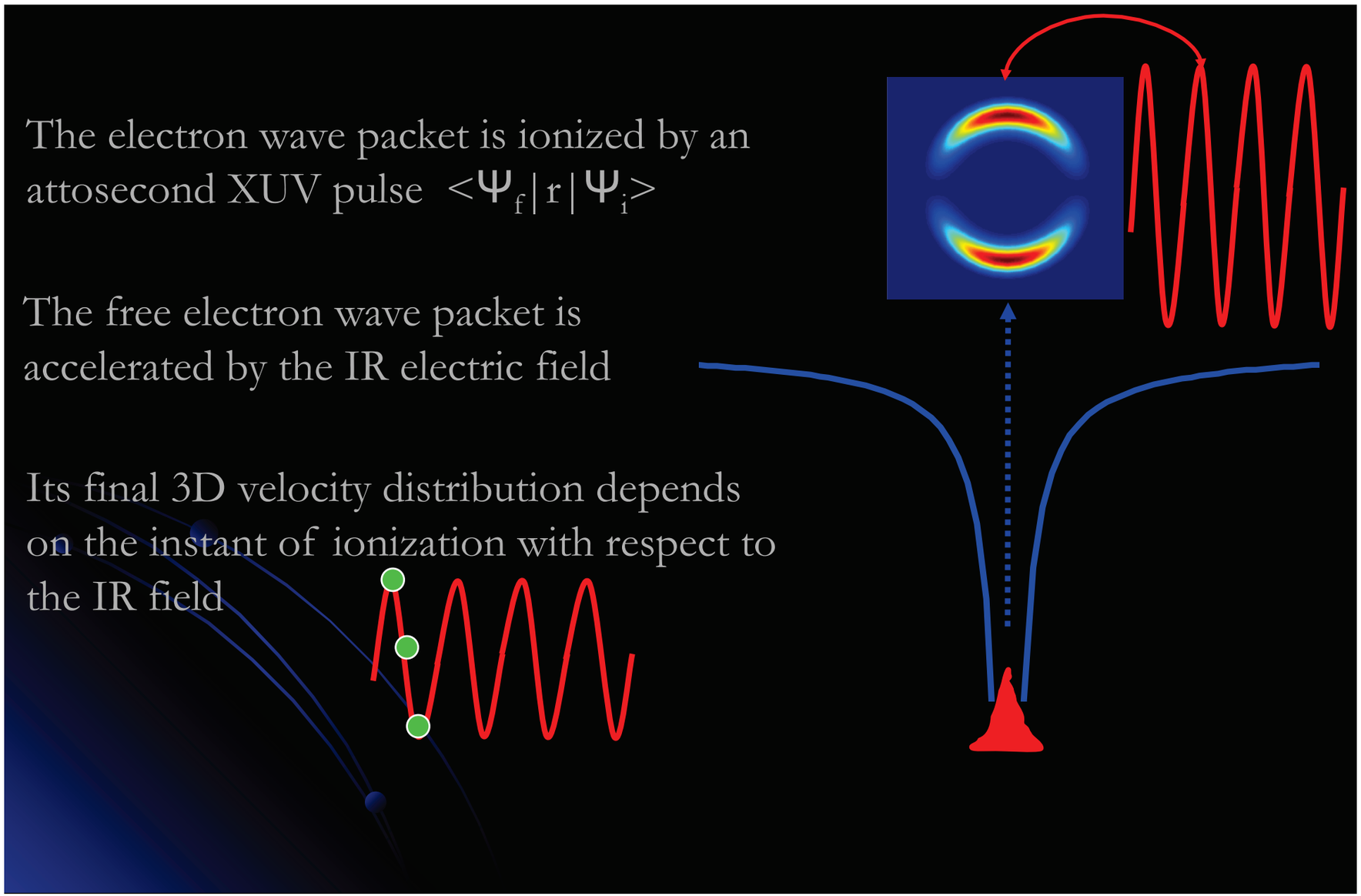
T. Remetter et al. ***Nature Physics*** *2, 353 (2006)*

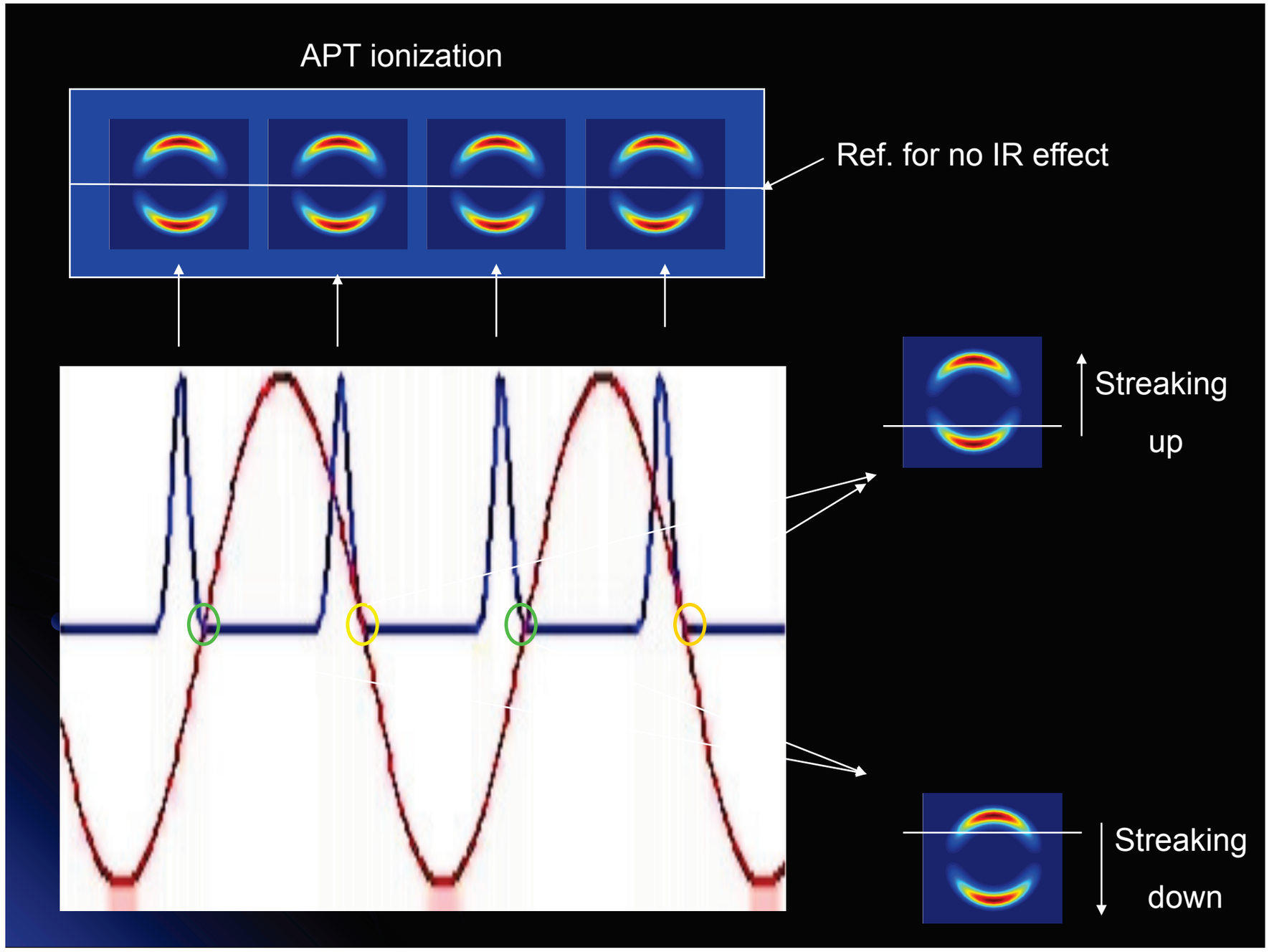
Streaking

The electron wave packet is ionized by an attosecond XUV pulse $\langle \Psi_f | r | \Psi_i \rangle$

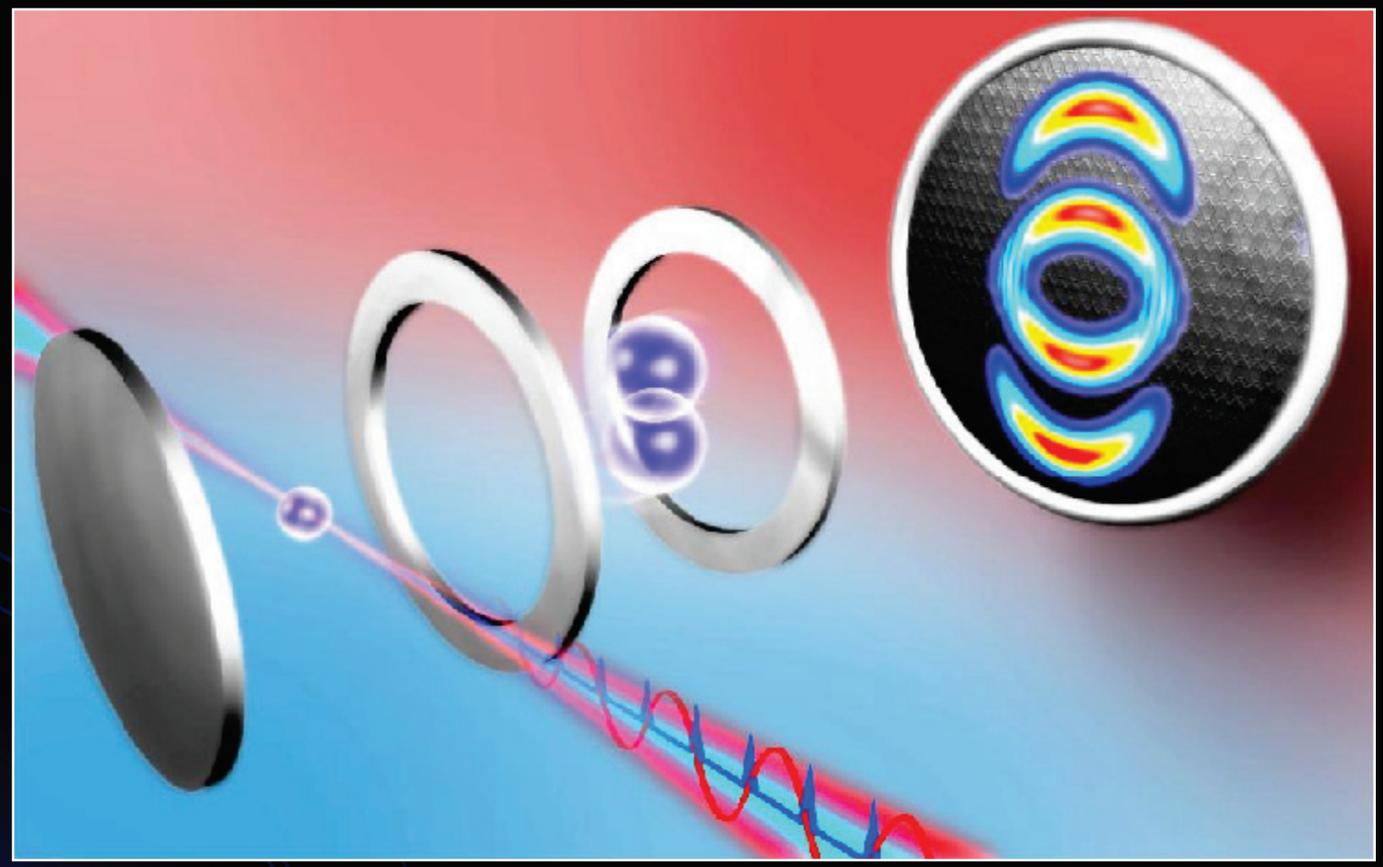
The free electron wave packet is accelerated by the IR electric field

Its final 3D velocity distribution depends on the instant of ionization with respect to the IR field

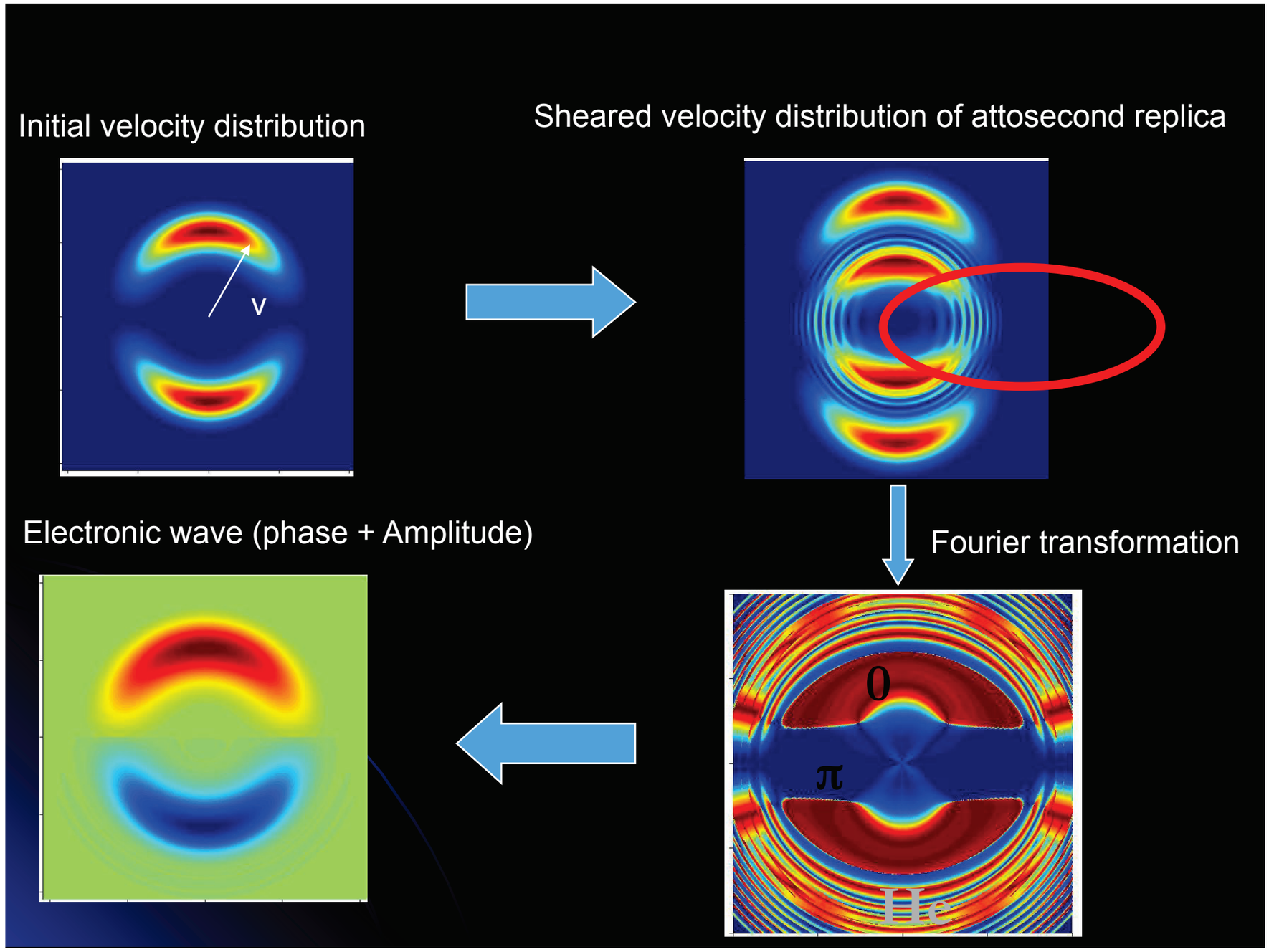



APT ionization
Ref. for no IR effect
Streaking
up
Streaking
down

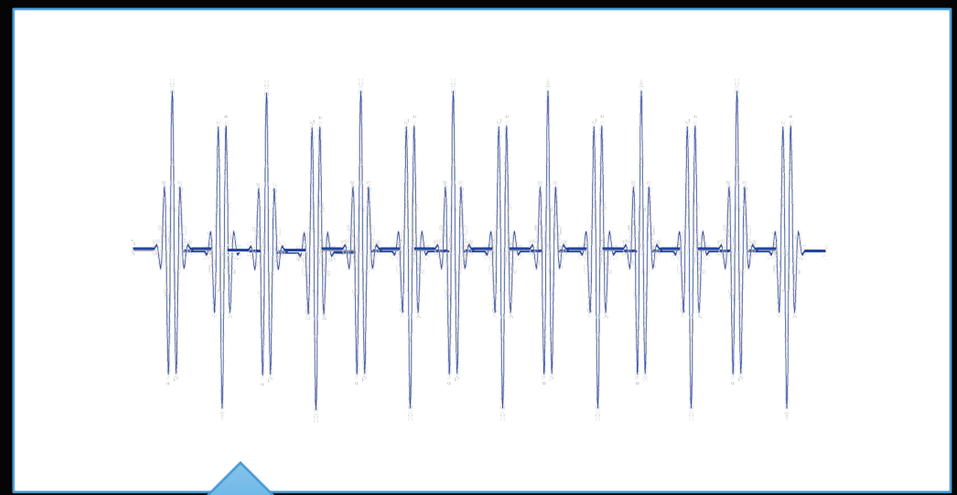
Shearing interferometer



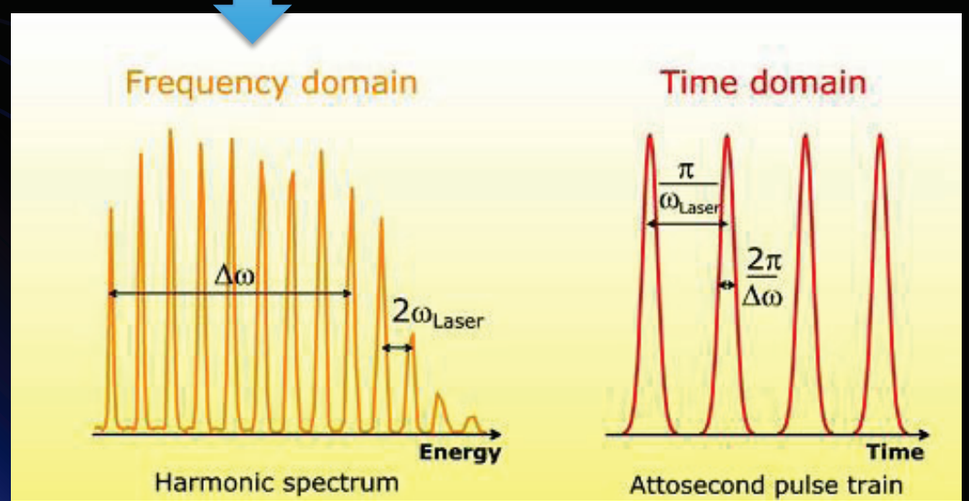
T. Remetter et al. ***Nature Physics*** *2, 353 (2006)*


Initial velocity distribution
v
Sheared velocity distribution of attosecond replica
Fourier transformation
0
π
He
Electronic wave (phase + Amplitude)

Rabbit

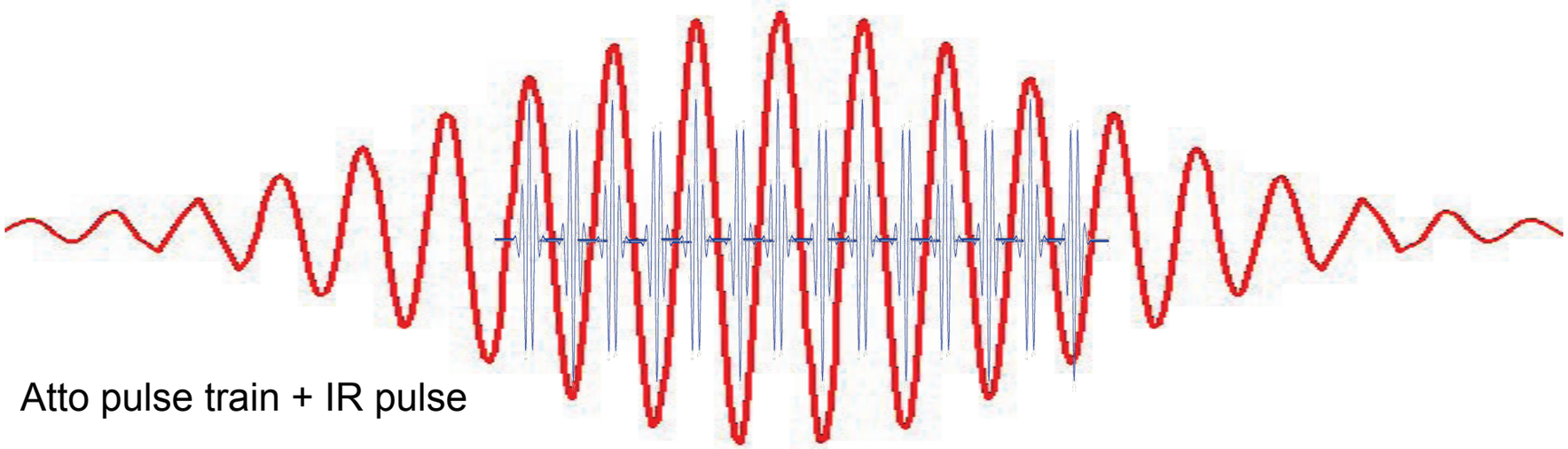


Atto pulse train

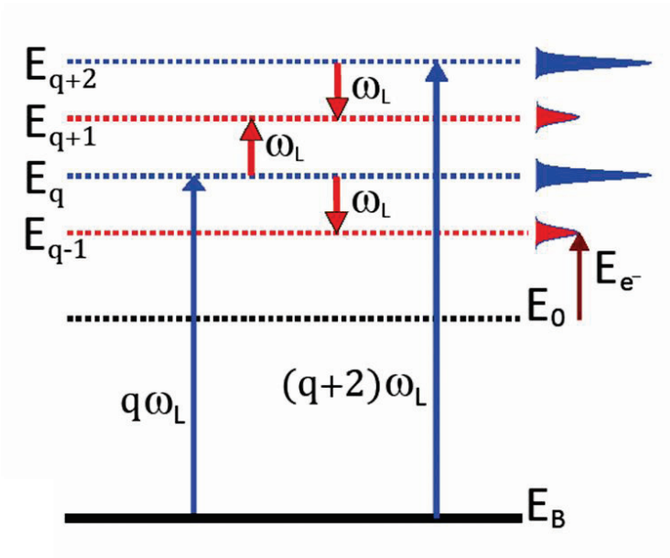


harmonic comb =
discrete photon energy

Rabbit



Atto pulse train + IR pulse

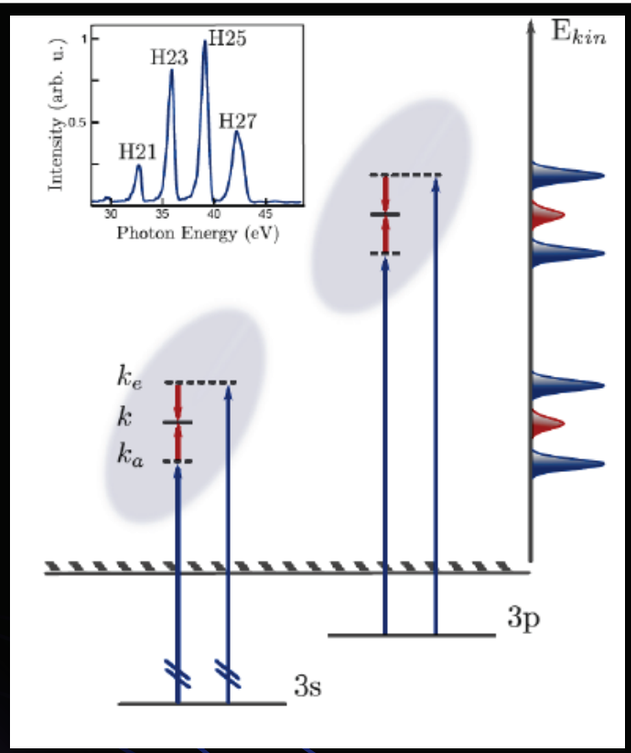


$S = a + b.\cos(2w_L * \tau + \Delta\theta - \Delta\varphi)$

- measure of the phase between harmonics

- measure of the atomic phase

Rabbit



How does it take for one electron to reach the continuum?
What does it depend on?

$$\hbar \frac{d\eta}{d\epsilon} = \tau_W$$

Scattering phase

Wigner time delay

Group of A. L'Huillier

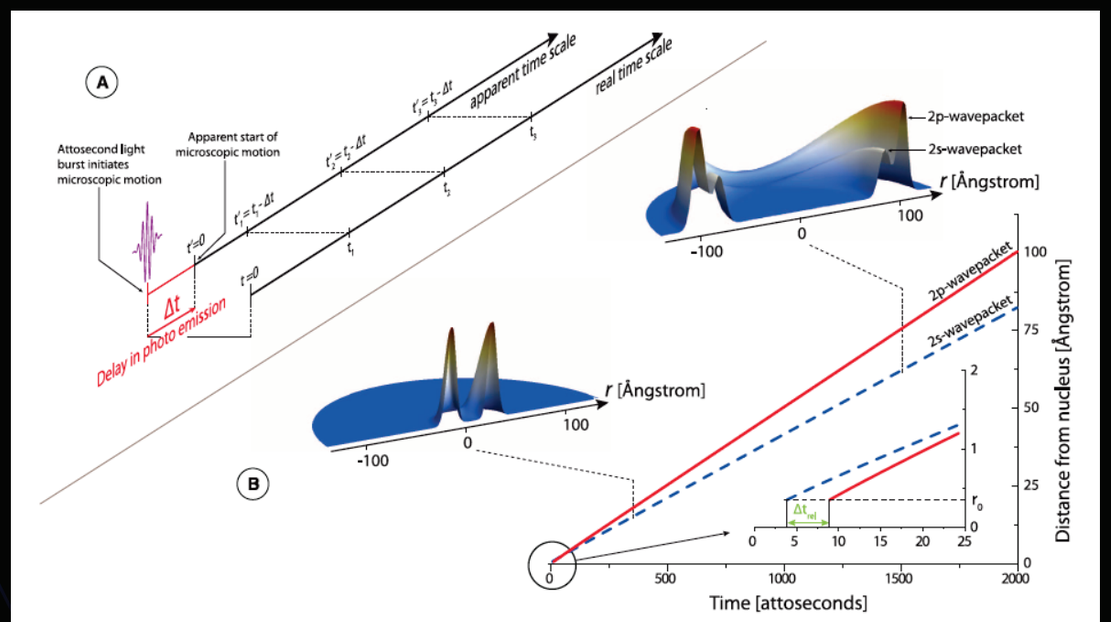
$$\cos(2\omega t + \Delta\phi_{harm} + \Delta\eta(3p) + \Delta\phi_{cc}(3p))$$

$$\cos(2\omega t + \Delta\phi_{harm} + \Delta\eta(3s) + \Delta\phi_{cc}(3s))$$

Shift!

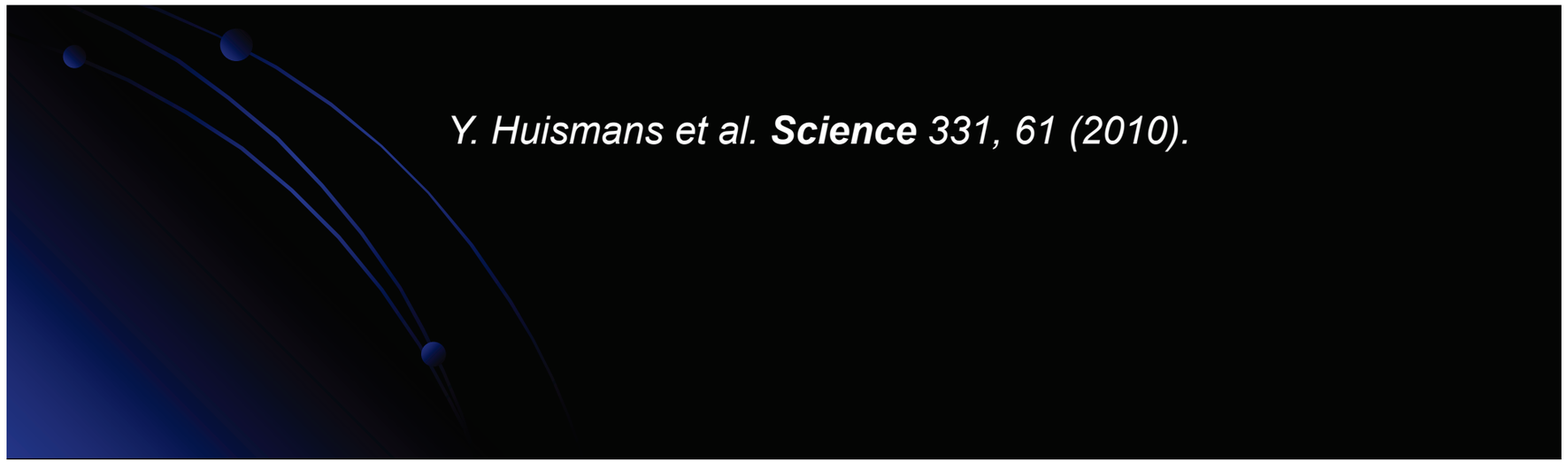
-V- Delay in photoemission

How does it take for one electron to reach the continuum?
What does it depend on?



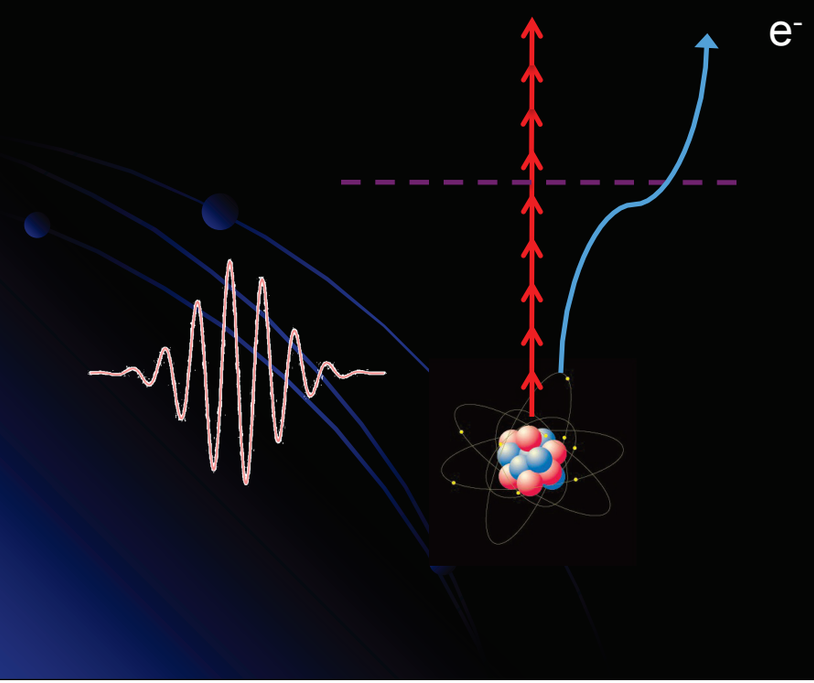
Science ***328, 1658 (2010);***
M. Schultze, *et al.*

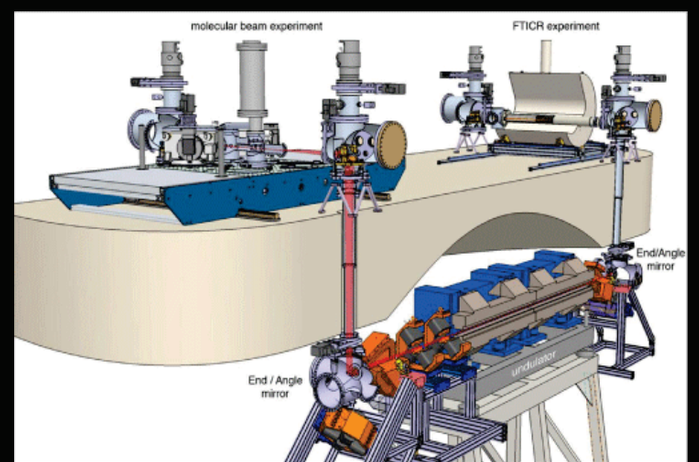
-VI- Photoelectron holography: Strong field ionization of Xe atom



-VI- Photoelectron holography: Strong field ionization of Xe atom

Strong field ionization of Xe atoms using far IR radiation at FELICE



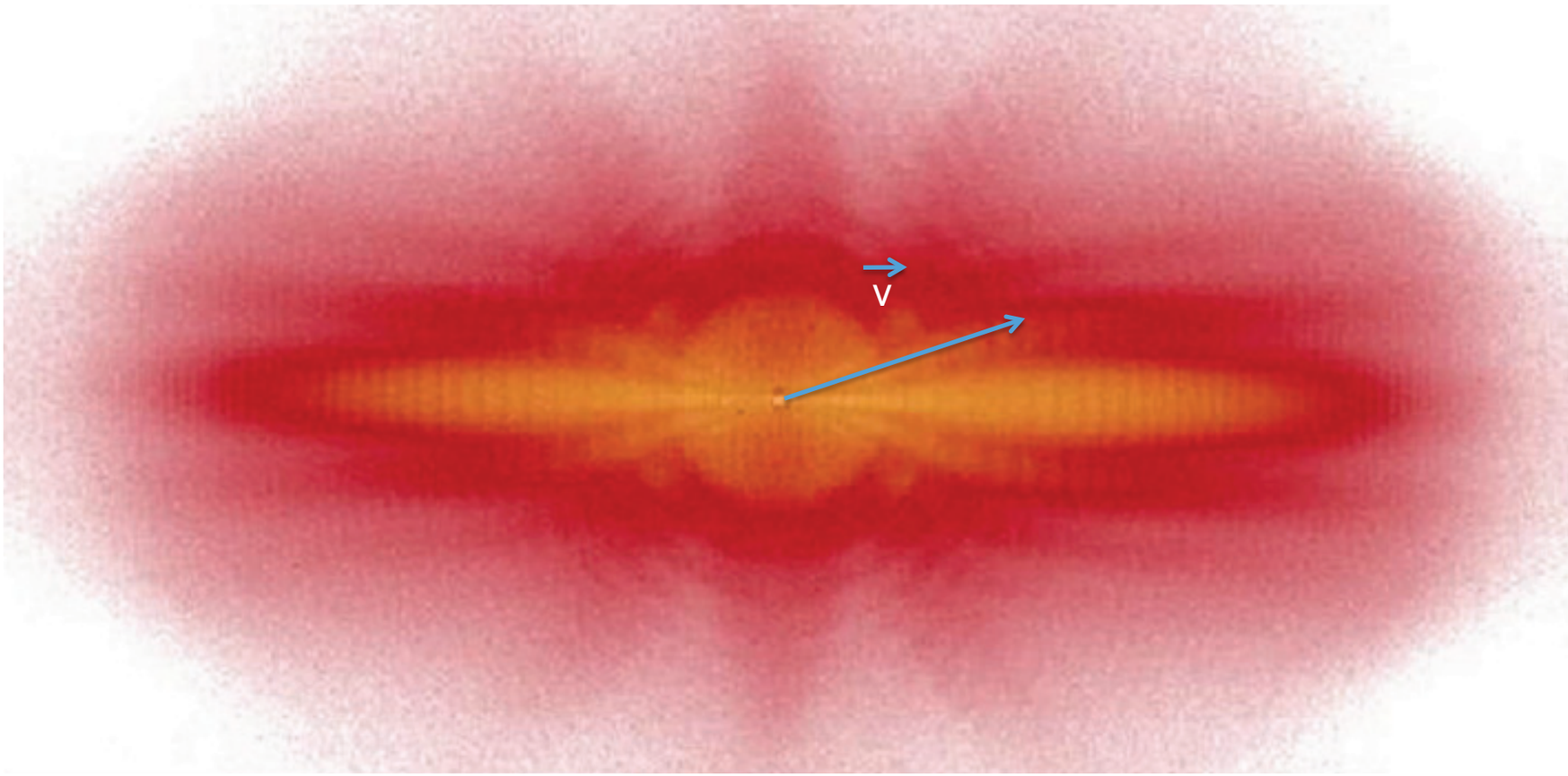


FELICE user's facility (The Netherlands)

1ps
5 μm
10^{12} W/cm^2

-VI- Photoelectron holography: Strong field ionization of Xe atom

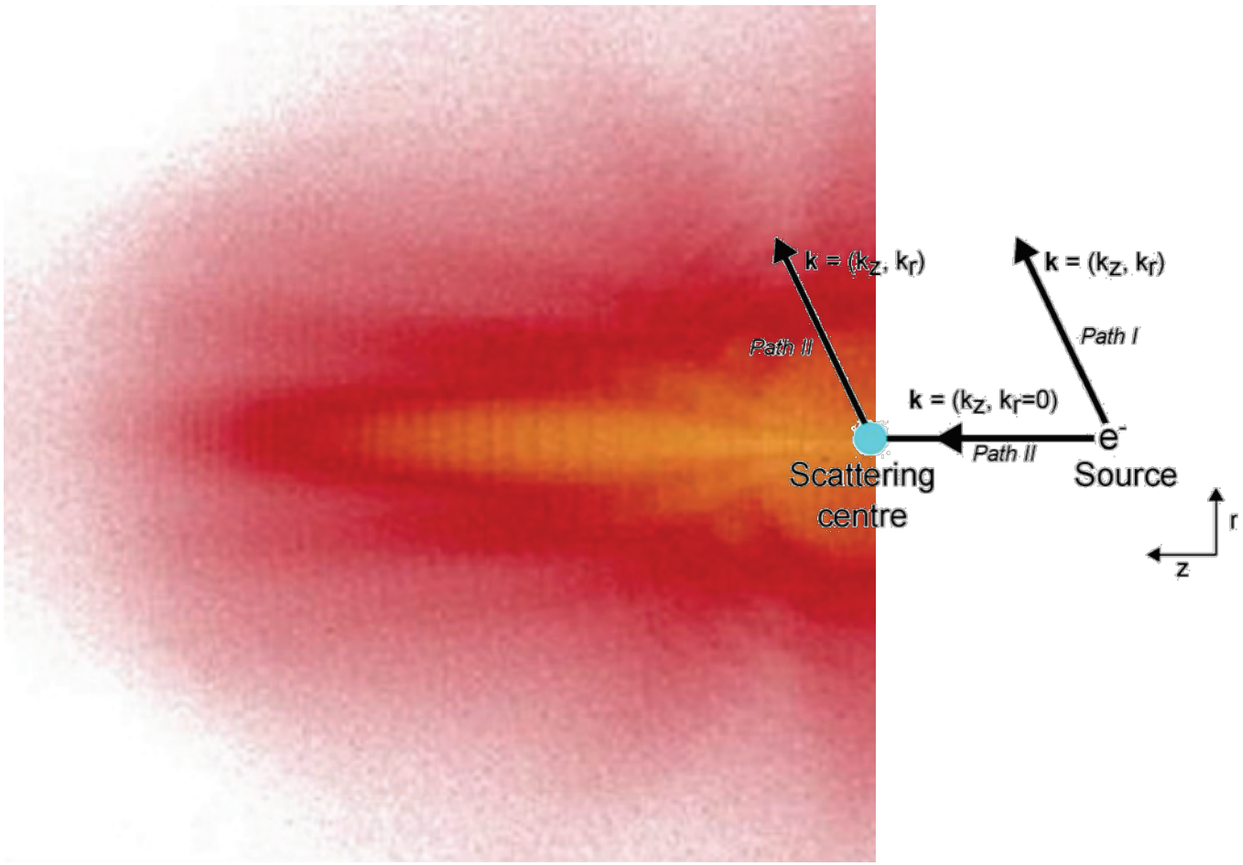
Y. Huismans et al. ***Science*** *331, 61 (2010).*



Measured electron momentum distribution => highly structured

-VI- Photoelectron holography: Strong field ionization of Xe atom

Y. Huismans et al. ***Science*** *331, 61 (2010).*



measured electron momentum distribution

-VI- Photoelectron holography: Strong field ionization of Xe atom

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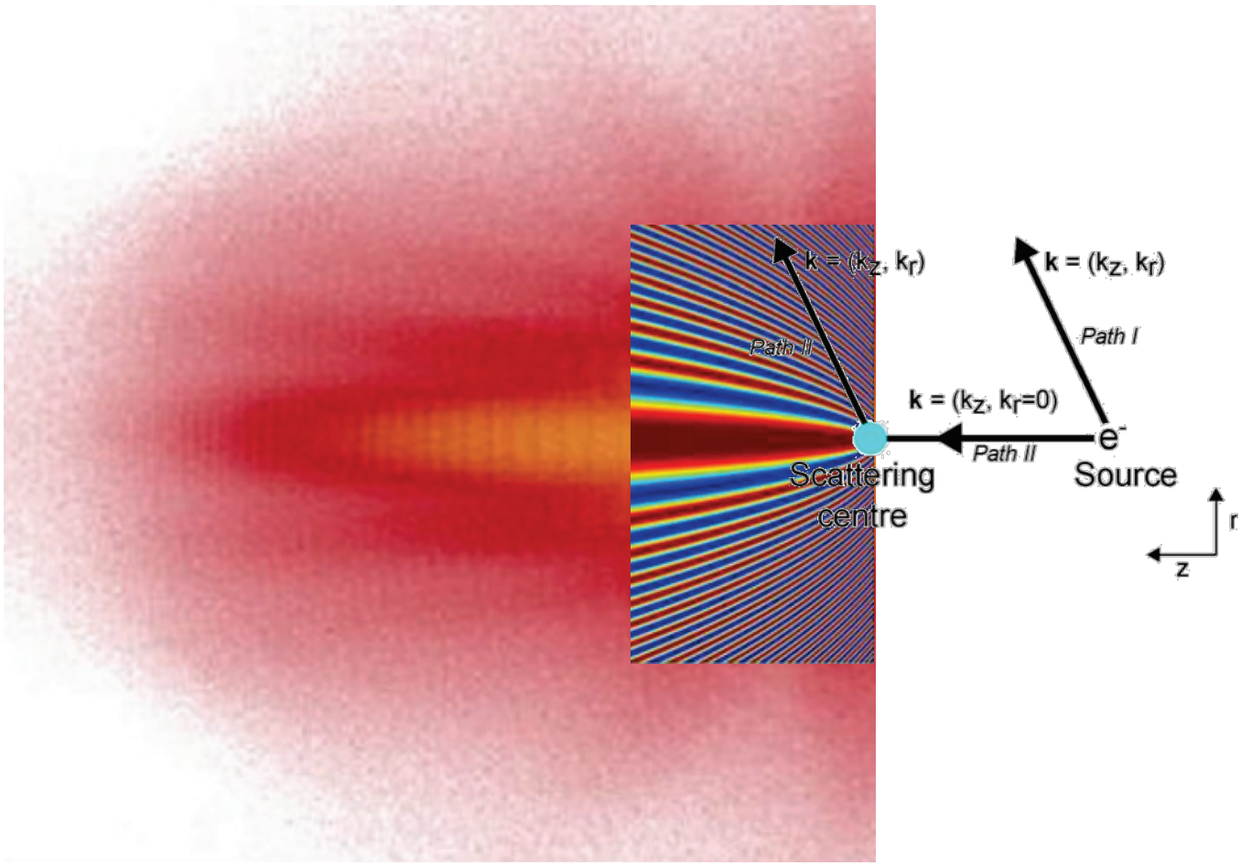
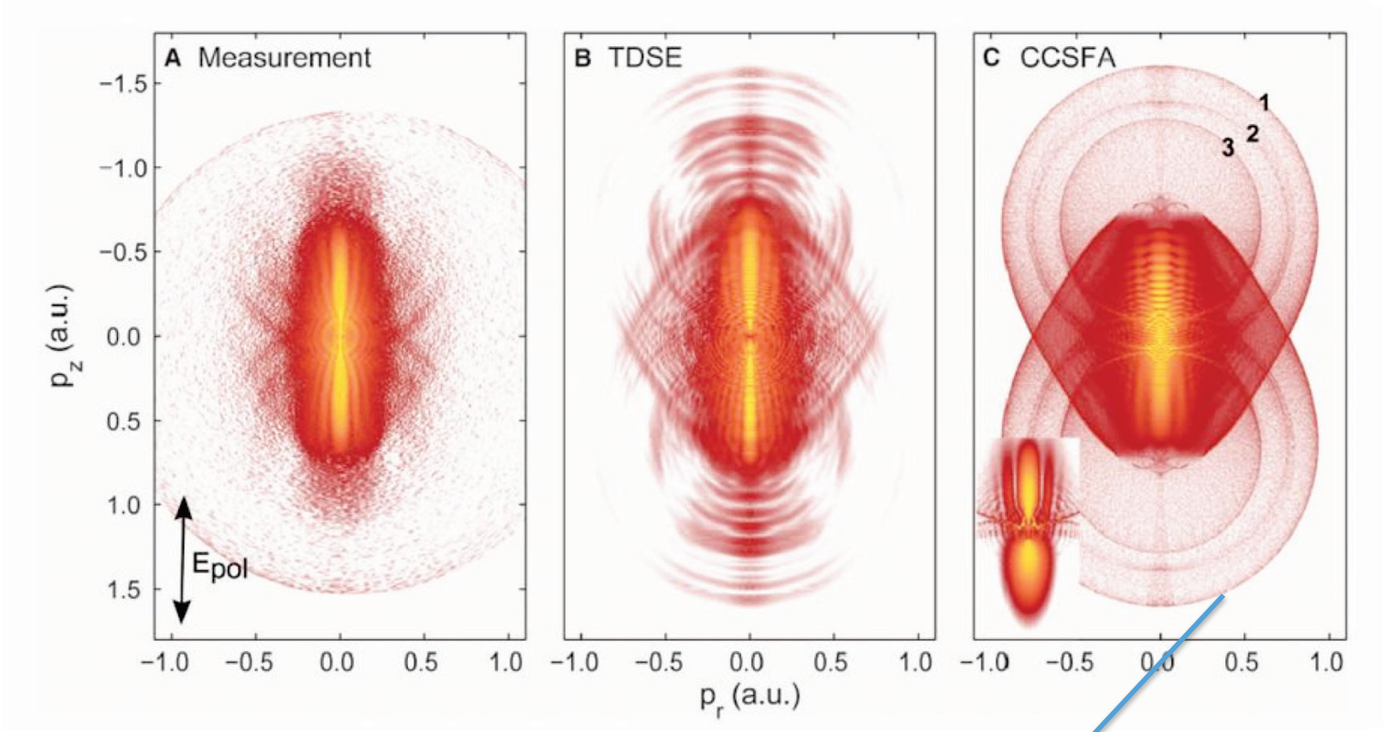


measured electron momentum distribution

-VI- Photoelectron holography: Strong field ionization of Xe atom



+ time resolved electron dynamics

Conclusion

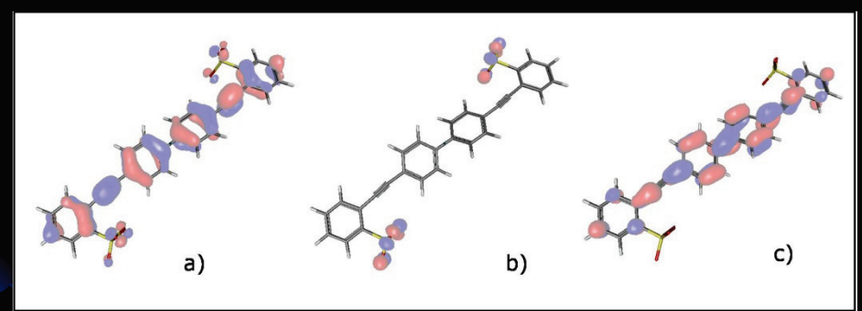
- FEL and attosecond science is highly relevant to study molecules and is at its infancy

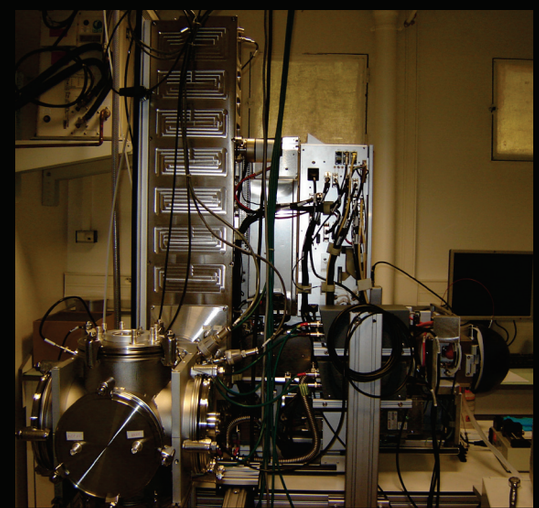
- First experimental demonstrations have been made on simple diatomic molecules

- Broad applications:
towards attosecond control, attochemistry, molecular electronics etc…

Conclusion

- Work on large polyatomic systems in progress (biomolecules, clusters etc …)





THERE IS A HUGE NEED FOR THEORETICAL INVESTIGATIONS …

TD-DFT has certainly a major role to play …

… calculate photoelectron, photoion spectra, dissociation, TD-absorption, nuclear motion (beyond herenfest?) etc…

Co-Workers

LASIM-CNRS Lyon

Thomas Barillot (PhD)
Cécilia Cauchy (PhD end sept 2011)
A. Marciniak (Master stud)
V. depré (Master stud)
A.R Allouche
G. Celep

LPMCN-CNRS Lyon

Group of M. Marques

LPT Toulouse
Group of E. Suraud

MaxBorn Institut (Berlin)

Group of M.J.J. Vrakking.

Milano
G. Sansone, M. Nisoli

LLC Lund
Group of Anne L'Huillier

Synchrotron Max lab
Group of S. Sorensen

FELICE (Rinhuizen)
J. Bakker, B. Redlich, L. van der Meer